

### U. S. DEPARTMENT OF AGRICULTURE,

BUREAU OF CHEMISTRY-BULLETIN No. 147.

H. W. WILEY, CHIEF OF BUREAU.

# COAL-TAR COLORS USED IN FOOD PRODUCTS.

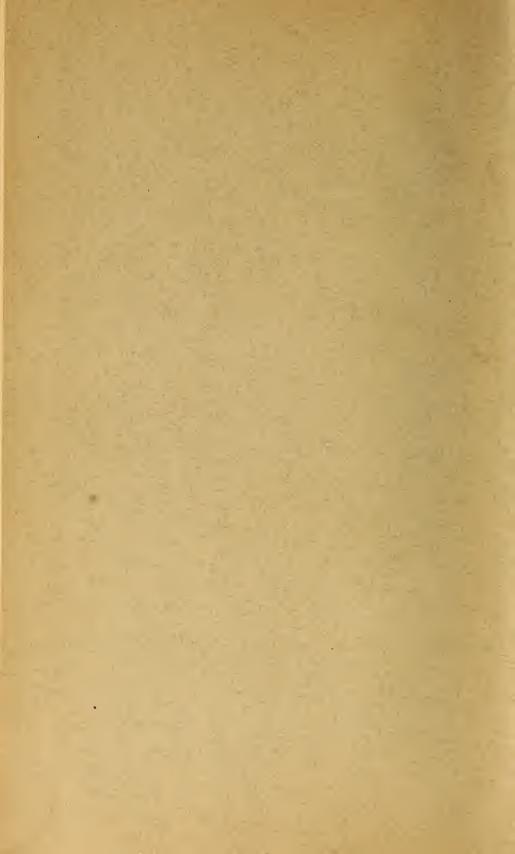
BY

BERNHARD C. HESSE, PH. D.

Expert, Bureau Chemistry.



WASHINGTON:
GOVERNMENT PRINTING OFFICE.
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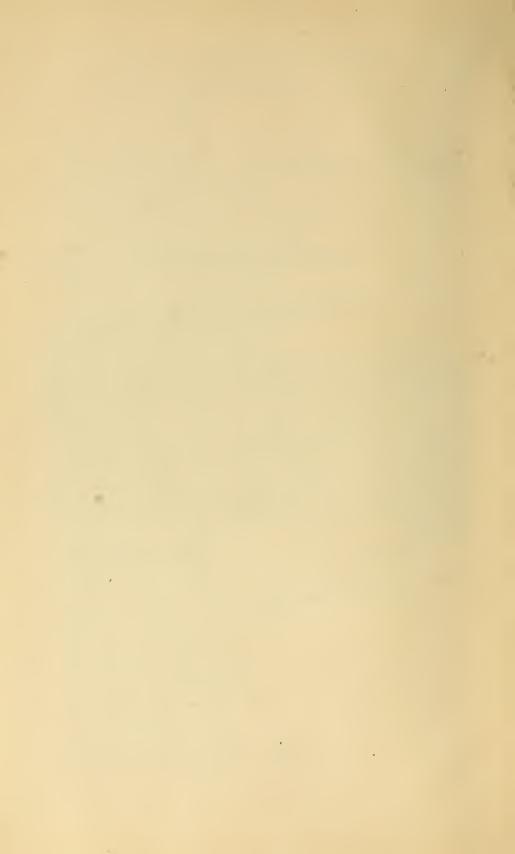
U. S. DEPARTMENT OF AGRICULTURE,
BUREAU OF CHEMISTRY,
Washington, D. C., January 31, 1911.

Sir: I have the honor to transmit for your approval a report by Bernhard C. Hesse, a color expert, containing both original chemical work done in the bureau since the passage of the food law on anilin dyes used for foods, and a valuable and extensive compilation of the literature of the subject, especially with reference to the harmfulness of coal-tar colors and their physiological effects. These data formed the basis of the opinions stated in Food Inspection Decisions 76, 77, and 106, and are presented in detail as of scientific and practical interest to all those concerned in the use of coal-tar colors in foods, whether as manufacturers, food officials, or consumers. I recommend the publication of this report as Bulletin No. 147 of the Bureau of Chemistry.

Respectfully,

H. W. WILEY, Chief.

Hon. James Wilson, Secretary of Agriculture.



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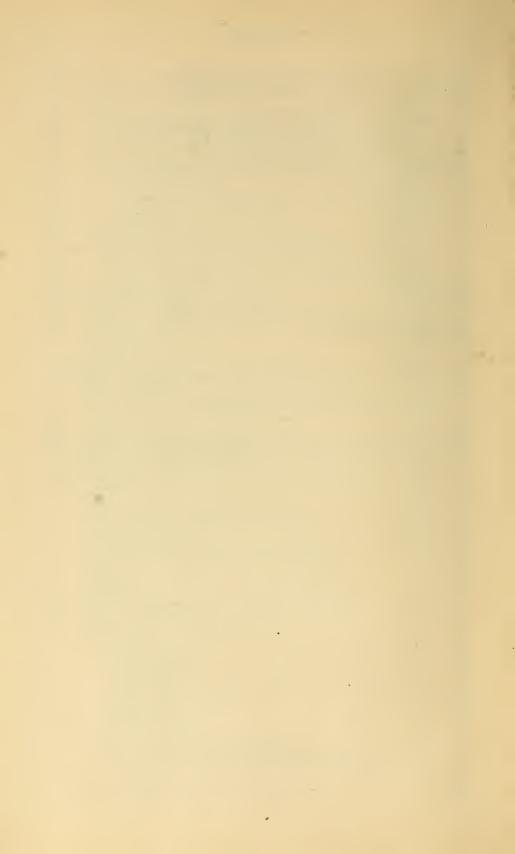
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## COAL-TAR COLORS USED IN FOOD PRODUCTS.

#### INTRODUCTION.

#### PURPOSE OF THE INVESTIGATION.

For the purposes of the investigation reported in the following pages, the legitimacy of the coloring of food and food products under certain conditions is regarded as established; the ethical and dietetic aspects of the question of food coloring are not here considered.

The means at hand for coloring food products may be conveniently classified as vegetable, animal, mineral or inorganic, and synthetic or so-called coal-tar colors or dyes. Representatives of each of these have at one time or another all been used in the coloring of food, and the laws of various European and American States have, from time to time prohibited the use of certain specified members or all of each or some of the foregoing classes. It is therefore obvious that even for the legitimate purposes for which food can be colored, improper means are at command, and some of these, if not all, have been prohibited by law at some time or another.

It is the function of the present work to determine what members of the synthetic or coal-car colors should be considered legitimate for coloring foods. It is confidently believed that the material collected in the following pages points clearly and solely to the following conclusions:

- 1. Coal-tar dyes should not be used indiscriminately in foods.
- 2. Only specified coal-tar dyes should be used in foods.
- 3. Only tested and certified dyes should be used in foods.

The work here reported has furnished the basis for Food Inspection Decisions Nos. 76, 77, and 106, issued July 13, 1907, September 25, 1907, and March 25, 1909, respectively, and the investigation itself was practically terminated January 1, 1910.

The effect of these decisions has been to restrict the coal-tar colors permitted for use in foods to seven specified and enumerated colors, until such time as it shall be shown with reasonable conclusiveness that other colors should be added to such list; and further, all coal-tar colors permitted for use in food are to be of a degree of purity and cleanliness acceptable to the Department of Agriculture, and are to be so certified.

In order to avoid any uncertainty as to the chemical composition of the enumerated colors, direct reference is made in Food Inspection Decision No. 76 to a standard work in which such chemical composition is clearly and unequivocally set forth. The relevant parts of Food Inspection Decision No. 76 are as follows:

The use of any dye, harmless or otherwise, to color or stain a food in a manner whereby damage or inferiority is concealed is specifically prohibited by law. The use in food for any purpose of any mineral dye or any coal-tar dye, except those coal-tar dyes hereinafter listed, will be grounds for prosecution. Pending further investigations now under way and the announcement thereof, the coal-tar dyes hereinafter named, made specifically for use in foods, and which bear a guaranty from the manufacturer that they are free from subsidiary products and represent the actual substance the name of which they bear, may be used in foods. In every case a certificate that the dye in question has been tested by competent experts and found to be free from harmful constituents must be filed with the Secretary of Agriculture and approved by him.

The following coal-tar dyes which may be used in this manner are given numbers, the numbers preceding the names referring to the number of the dye in question as listed in A. G. Green's edition of the Schultz-Julius Systematic Survey of the Organic Coloring Matters, published in 1904.

The list is as follows:

Red shades: 107. Amaranth. 56. Ponceau 3 R. 517. Erythrosin.

Orange shade: 85. Orange I.

Yellow shade: 4. Naphthol Yellow S.

Green shade: 435. Light Green S F Yellowish.

Blue shade: 692. Indigo disulfoacid.

Each of these colors shall be free from any coloring matter other than the one specified and shall not contain any contamination due to imperfect or incomplete manufacture.

The reasons, broadly considered, which led up to these food inspection decisions are given in concise fashion in this introduction.

Looking over the restrictions placed upon coal-tar colors by the lawmakers of the various countries it will be found that certain colors are in some instances specifically prohibited and in other instances that certain specific colors, or classes of colors, and only such, are permitted for the legitimate purposes of food coloring.

Private organizations, such as the Swiss Society of Analytical Chemists and the National Confectioners' Association in the United States, have also made recommendations permitting specific colors only, and in addition specifically prohibiting others. Individual authors have likewise made similar recommendations. The control of the quality of the food colors practiced on the part of those Governments which restrict the use of coal-tar colors to certain individuals, so far as any publications show, has not been very extensive.

The action taken against the use of coal-tar colors for food-coloring purposes has ranged all the way from absolute prohibition of their use for any purpose whatsoever to the practically unlimited use in legitimate food coloring operations of all but two of such colors.

Intermediate between these two extremes we find the prohibition of a greater number than two, or of all the members of this class except certain specified colors, and even here with the restriction that they shall be used only for certain legitimate food-coloring purposes.

It would be desirable to have a number of coal-tar colors of established harmlessness specifically permitted, particularly if the number be sufficient to meet all the legitimate demands arising in the foodcoloring art. To prohibit only specified coal-tar colors and, by implication, to permit all the rest of this class, would allow the unrestricted use of newly discovered colors, and all other coal-tar colors not examined as to their effect on health. A limited list of permitted coal-tar colors which would make the use of all coal-tar colors outside of the permitted list illegal would properly protect the health and could work no substantial hardship upon those engaged in food coloring. Any such hardship would be avoided by providing that if it is shown that none of the colors of the permitted list meets certain legitimate requirements and that coal-tar colors outside the permitted list are capable of satisfying this need and are in and of themselves harmless the permitted list can be expanded by the proper authorities to meet additional needs or growing requirements without exposing the public health to any risk.

#### NUMBER OF COLORS PERMITTED.

It will be shown in the following pages that in the summer of 1907 there were on the market of the United States 80 different chemical individuals, or so-called coal-tar colors, offered for the coloring of food. It has been known since 1888 that it is unsafe to attempt to predict the harmfulness or the harmlessness of coal-tar colors by inference or analogy; therefore an ideally perfect permitted list should contain only such colors as have each been examined physiologically, separately, and specifically, and their harmlessness determined by actual test. Out of the 80 colors referred to 30 had not been examined at all, so far as the literature shows, and therefore their harmlessness is certainly open to question; 26 had been examined physiologically, and the published accounts with respect to their harmlessness or their harmfulness are in each case contradictory; on 8 none but adverse reports were to be found in literature, leaving only 16 out of 80 colors on the market which had been established with more or less certainty as harmless—that is, the users of these 8 colors were deliberately taking chances with the public health, since the harmful nature of those 8 had for a long time past been known to those conversant with such subjects; the use of the 26 doubtful colors is more defensible than the use of the 8 known to be harmful. Out of the 30 of whose action nothing was known it can not be said

how many are or are not harmful, nor can the risk forced upon the public health be satisfactorily measured.

This brief summary must suffice for the present as a justification for the restrictions of the permitted colors to 7 in number. The full reasons for each and every step will appear in their proper places in the pages following.

#### QUALITY AND EFFICIENCY OF COLORS PERMITTED.

An examination of 30 specimens representative of the 7 selected permitted colors on the United States market in the summer of 1907 disclosed such a condition of uncleanliness of product, or careless or improper manufacture, and the use of such utterly inferior qualities of products for food coloring purposes, that control over the quality of the seven permitted colors seemed necessary. The results of the work in the making and maintaining of standards of quality for each of these seven colors also justify this control. there was in 1907, and for a year or more later, a considerable divergence of opinion among chemists as to what should be the proper quality requirements for these colors is shown by the fact that out of 72 foundation certificates offered in accordance with Food Inspection Decisions Nos. 76 and 77, 57 were rejected on their face because they did not comply with the standards of quality then in mind, or then shown to be commercially attainable. Much objection has been made by many of those whose certificates were rejected on the ground that the standards then in mind were unreasonable, unjustifiable, and nonattainable. The actual results, however, are that with very few exceptions the standards in mind early in the work have all been exceeded in practice; the 41,000 pounds (20.5 tons) of certified colors now in existence made in 97 batches, or an average of more than 420 pounds per batch, are, with the exception of perhaps one or two first batches, far cleaner than was expected when the 57 certificates above referred to were rejected.

There has been no complaint against the permitted colors for want of efficiency or for the possession of unsuitable attributes, which has been pressed or sustained with any such earnestness as would reasonably be expected if the defects complained of were as great as they were represented. Complaints have been made against the yellow, when used in acidulated fruit sirups, on account of its possessing a bitter taste; the proof of this, so far as any has been offered, was for a long time not of a convincing nature, and it was two years after the first objection was raised before any concerted or positive action was taken by those interested. The yellow has also been criticized because it is not sufficiently fast to light; although it was satisfactorily shown that another yellow was faster to light than the permitted yellow, no one has maintained that the yellow desired

was actually preferable to the permitted yellow wholly and solely because of its superior fastness to light.

The blue has been criticized because it is not of the proper shade to permit of its use in the bluing of sugar, but the substitute offered therefor has not been supported by its sponsors in a way to indicate that a defect of serious magnitude exists. On the grounds of sufficiency and of efficiency the list of permitted colors selected appears to have been justified by the absence of any real or substantial complaint against them, on either or both of these grounds, during a period of more than three years.

None of the seven permitted colors is patented; their manufacture and their purification are open to all, and none of the 80 colors on the market in the summer of 1907, with perhaps one exception, had been discovered since 1891; in other words, the advances in the coaltar industry from 1891 to 1907 had added nothing to the colors

serviceable to the art of food coloring.

The list of colors permitted in Food Inspection Decision No. 76 embraces, therefore, a sufficient number of colors for all legitimate food-coloring purposes, the coloring of fats, oils, butter, etc., excepted, for which no suitable color had been examined and reported in the literature as being harmless and fit for use in foods; they can be made by any one; no one can have a monopoly in any one of them by virtue of patents; any competent maker can make all or any of them and purify them to the required degree of cleanliness. The standards growing out of the control exercised by the Department of Agriculture are such as to insure that the colors used for food-coloring purposes possess a proper degree of cleanliness and such a degree of cleanliness is commercially feasible and is a commercial reality.

The policy adopted in this respect is therefore justified not only from the viewpoint of the history of the attempts on the part of various governments to control the quality of food colors, but also by the results actually obtained by its adoption. This policy of restricting food colors to certain chemical individuals and demanding that those possess certain qualities is in complete harmony with the following suggestion made to the commission on rules and regulations under the food and drugs act, at its hearing in New York, in September, 1906.

Any kind of a harmless color should be permitted, provided it is not a color generally known to be poisonous, or generally found to be poisonous, or one that may be almost impossible to be produced without containing some poison within itself when finished and ready for use. Coal-tar colors, as a class, should not be prohibited, but all those coal-tar colors generally found to be poisonous, or which are hard to produce without containing some poisonous properties when ready for use, should be forbidden the privilege of being used, or offered for sale for use in food.

Under the provisions of section 2, we have this to recommend to the commission, that every person selling or using a coal-tar color in food or drink, should be required

to secure, either on his own account, or from the person from whom he buys such color, a certificate to the effect that the identical color used has been tested for poisonous ingredients, and is, to the knowledge of the chemist making the test, absolutely harmless. The chemist should be required to be a competent physiological chemist, and must certify as above under oath. This would mean not that each package of color would have to be tested, but that every batch would have to be tested, and the certificate would then be held to relate to every batch. Such tests should be made in the United States and the chemist certifying should reside in and be a citizen of the United States.

It should not be deemed sufficient to have any particular brand of coal-tar color tested once, and a blanket certificate given, covering the whole brand as long as it may be sold, but every ounce of coal-tar color put out by a color manufacturer should be shown by actual test to be harmless.

While the exact mode of reaching the end in view is somewhat different from the one suggested above, yet the fundamental object, that each batch of color used in foods shall be specifically tested, and that such colors shall be harmless, is attained with reasonable certainty; and although there are colors other than these seven which are undoubtedly equally as little objectionable, and while it is true that the present policy contemplates the permitted use of but seven specific colors, yet that policy, as before outlined, is sufficiently broad and elastic to enable the addition of a color to the permitted list, when it is shown that such color really fills a need, not properly satisfied by one of the colors already permitted or some combination of these, and is in and of itself harmless. There can be no objection to the expansion of the list to such an extent as to include every harmless coal-tar color in existence; but the burden of proving such real need and harmlessness is very properly placed upon those who are seeking such expansion.

As far back as 1892 the following statement was made on page IV of the Leffmann translation of Weyl's book on coal-tar colors, in connection with the various European legislative enactments: "It is certain that none of these plans is even approximately satisfactory and the problem will be even more difficult of solution in the United States; indeed, it seems to me to be unsolvable." In view of this opinion the results of the food inspection decisions as herein shown may properly be regarded as, at least, a step in the right direction toward the solution of this problem.

This opinion is further supported by C. A. Neufeld who, in reviewing Food Inspection Decision No. 76, says: "The idea of permitting only specific selected coloring matters for use in the production of articles of food, and of excluding all other colors from such uses, must, in the interest of control of articles of food, be regarded as an extraordinarily happy one; a similar regulation is to be urgently recommended for our own country." (Zts. Nahr. Genussm., 1908, v. 15, p. 434.)

#### IDENTITY OF COAL-TAR COLORS USED IN FOOD PRODUCTS IN THE UNITED STATES IN 1907.

#### COLLECTION OF SAMPLES.

The question "Which coal-tar colors shall be permitted for use in coloring such food products as are to be consumed within the United States?" can be answered, "All colors now in use, or to be used for that purpose, provided they are harmless and necessary as defined on page 14."

This involves the further questions:

1. Which coal-tar colors of the 695 different chemical individuals now on the world's markets are actually used in the United States for

that purpose?

2. If restricted to such coal-tar colors as are now in use in the United States for this purpose, would this be likely to hamper or interfere with the invention of other coal-tar colors suitable for the coloring of food?

It would be physically impossible to go to every user of coal-tar colors in food products in the United States and obtain specimens of the coal-tar colors so employed; this would be impracticable not only because of the large number of such users, and their wide geographical distribution, but also because they often do not know what they are using, and further because of a reluctance, undoubtedly to be encountered among many, to disclose the nature of the products employed. This is rendered more than likely by the attitude of some of the makers of coal-tar colors, or their accredited agents, as will be shown later.

However, the sources of coal-tar colors are limited in number. By reference to pages IX and X of "A systematic survey of the organic coloring matters," by Arthur G. Green, published in London and New York by Macmillan & Co. (Ltd.), in 1904 (hereinafter referred to as "Green Tables"), it will be seen that there are approximately 37 different concerns the world over engaged in the manufacture of coal-Therefore a canvass of these sources for such coal-tar tar colors. colors as in their judgment, or in their business practice, they regard as proper for use in food products, is the best way of arriving at a fair demarcation of the field of coal-tar colors here in question.

Communication was therefore had with 13 actual manufacturers of coal-tar colors, in an endeavor to obtain from them such coal-tar colors as in their judgment or business practice are suitable for use, or are used in food products.

A request was also made for information as to the chemical composition of the coal-tar color specimens submitted; in order to avoid confusion, it was further asked that reference be made to the Green Tables, in which each chemical individual or coal-tar color has its own number, and if any of the contributed specimens was not so listed that the chemical composition be stated in a manner analogous to that used in the Green Tables. This procedure was necessary in order to reduce the terminology to a common and nonequivocal basis. Out of the 13 makers, or their accredited sole importers or selling agents in the United States, who were consulted, 9 have supplied the specimens requested; the remaining 4 promised to contribute, but have not done so. In the following table is shown the amount and character of the information obtained:

Tabulation of distribution of replies and character of information received.

	Ge	ographical	distributi	ion.	Manushan	Number	Number	No	
Country.	Coal-tar color makers.	Makers asked to send samples.	Makers not sending samples.	Makers sending samples.	Number of samples contri- buted.	of samples referred to Green Tables.	samples not referred to Green Tables.	composi- tion or ambigu- ous ter- minology.	
GermanyEnglandFrance	16 8 5 5	6 1 1	1	6 1	181 12	106 12	1	74	
Switzerland	5 1 1	3 2	3	2	61	35	5	21	
Total	37	13	4	9	254	153	6	95	

In order to make provision for the 24 makers listed in the Green Tables and not included in the 13 makers addressed requests for samples were sent to two domestic houses which import coal-tar colors from scources other than the above, for use in food products; their products must fairly represent any of the colors not covered by the 13 makers addressed. Of these two importers, one responded with 13 samples, and of each he gave the number in the Green Tables corresponding to each specimen; the other importer has not redeemed his promise to contribute specimens.

A third importer volunteered the Green Table numbers of four out of five coal-tar colors used in his business, but could not even approximately say what the remaining color was chemically. He did not contribute any specimens, nor was that necessary at the time this information was volunteered.

A fourth importer contributed specimens of five coal-tar colors needed in his business, but was able to give Green Table numbers for only three of them; he could not give even approximately the chemical composition of the remaining two.

Out of the 17 responsible concerns consulted 5, or 29 per cent, have not found it to their interest to contribute either specimens or information.

#### CLASSIFICATION OF SAMPLES SUBMITTED.

#### GREEN TABLE NUMBERS.

Out of the 284 specimens contributed, or reported on, 172 (60.6 per cent) were identified as to their chemical composition, by reference to the Green Tables; 6 (2.1 per cent) were otherwise unequivocally identified chemically, and for 106 (37.3 per cent) the makers, or their responsible agents, declined to state the chemical composition, i. e., 62.8 per cent were unequivocally identified, and the remaining 37.3 per cent were not so identified.

The specimens submitted are therefore divisible into the following

three classes:

Class I. Those for which numbers were given in the Green Tables, numbering 172.

Class II. Those whose composition was given in chemical language,

numbering 6.

Class III. Those whose composition was not given in any language capable of correct and certain translation into chemical terms, numbering 106.

Consider Class I. The Green Tables, page VI, divide the coal-tar colors into 21 groups, comprising 695 different chemical individual coal-tar colors. The 172 members of Class I number in all 74 individuals, or 10.6 per cent of the Green Tables, and fall into 11 of the 21 groups of those Tables.

The following table classifies the samples according to the Green Table groups:

Green Table groups and number of collected samples falling within them.

Color groups of the Green Tables.	Number of mem- bers in		samples into the groups.	Color groups of the Green Tables.	Number of mem- bers in	Collected samples falling into the several groups.		
	group.	Number.	Per cent.		group.	Number.	Per cent.	
Nitro Monoazo Disazo Trisazo	126 204 46	1 30 11	16 24 5	AnthraceneIndophenolAzin	37 8 38 32	2	5	
Tetrakisazo Nitroso Stilbene	11 5 18	1	20	Thiazin		$\frac{2}{1}$	22 25	
Oxyketone. Diphenylmethane. Triphenylmethane. Xanthene.	8 2 66 35	1 15 9	50 23 26	SulphidIndigo	21 7 695	1 74	15	
Acridin	6	9			090	74	10.0	

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#### SOURCE.

The distribution of the 74 different chemical individuals of Class I among the 12 different sources from which they were obtained is as follows:

Distribution of the 74 different samples of Class I among the 12 sources supplying same.

Number.	Per cent.	Number of sources from which each came.	Number.	Per cent.	Number of sources from which each came.
35 20 4 4 5 3	47.3 27 5.4 5.4 8.1 2.7	1 2 3 4 5 6	1 1 0 1 0 0	1. 35 1. 35 1. 35	7 8 9 10 -11 12

It follows from this table that there is very little unanimity among the different concerns furnishing coal-tar colors for use in food products as to which of their products are desirable, necessary, or suitable for such use.

Inspection of this table shows that only three colors out of 74, or 4 per cent, were wanted by more than half of all the sources; that only 6, or 8.1 per cent, were wanted by half of the sources; and that not one of the colors was wanted by all the sources. This last statement is true of manufacturers as well as importers, each group taken by itself.

#### PATENTS.

This lack of unanimity is not due to the patent situation, because not more than one of these 74 products is patented, and it is more than likely that the United States patent on this product has long since expired.

Moreover, only 6 of the 12 sources offered colors at one time patented by themselves or others. The total number of such expatented products is 45, and of these only 22 were offered by those who had patented them; the remaining 23 were offered by sources other than the ex-patentees, and were not offered by such ex-patentees.

Patented colors.

Total number of patented products offered.	Ex-patented products offered by patentee.	Total number of patented products offered.	Ex-patented products offered by patentee.
11 16 6 7	2 8 5 4	1 4 45	1 2 22

It would, therefore, seem to be rather clear that others think more favorably of such ex-patented products as food colors than do the original patentees. In view of the fact that the latter would generally be in a better position, and would have greater opportunity than any one else to judge of the suitability of the patented products for use in food products, it may well be inferred that such products are not altogether free from disadvantages as food colors.

The second of the two questions propounded, namely, If restricted to such coal-tar colors as are now in use in the United States for this purpose, would this be likely to hamper or interfere with the invention of further coal-tar colors suitable for the coloring of food products? can be answered "No" because none of the colors submitted was discovered later than 1891; out of the 214 coal-tar colors since then discovered not one was among those submitted for use in foods, and out of the 481 discovered in 1891 and prior thereto, only 74 were so submitted, or 2 out of every 13 of such colors. In the following table these data are given year by year:

Year of dis- covery.	Num- ber of sub- mitted colors discov- ered.	Total number of coaltar colors discovered.	Year of dis- covery.	Num- ber of sub- mitted colors discov- ered.	Total number of coaltar colors discovered.	Year of dis- covery.	Num- ber of sub- mitted colors discov- ered.	Total num- ber of coal- tar colors discov- ered.	Year of dis- covery.	Num- ber of sub- mitted colors discov- ered.	Total num- ber of coal- tar colors discov- ered.
1740 1856 1859 1861 1862 1863 1867	1 1 2 1 1 1	1 3 1 3 5 3 3	1871 1874 1875 1876 1877 1878 1879	1 1 6 5 2 12 7	6 4 12 9 10 26 23	1881 1882 1883 1884 1885 1886	4 5 9 3 2 3	11 20 25 16 21 32	1887 1888 1890 1891 Total.	1 1 1 3 74	29 36 46 33 378

#### SHADES OF COLOR.

The sufficiency of the 74 colors used for food-coloring purposes in the United States, for any and all tinctorial ranges, no matter how refined, appears from the following table:

Green Table numbers of the 74 submitted colors showing shades and number of sources supplying each.

	Nu	ımber	of sour	ces out	t of a p	ossible	12 offe	ering e	ach col	or.	Total
Shades.	1	2	3	4	5	6	7	8	9	10	of dyes offered for each shade.
Red	104 105 169 240 584	65 108 146 462			106	103	107				12

Green Table numbers of the 74 submitted colors showing shades and number of sources supplying each—Continued.

Shades.	Nı	ımber	of sou	ces ou	t of a p	ossible	e 12 off	ering e	ach co	lor.	Total number of dyes
	1	2	3	4	5	6	7	8	9	10	offered for each shade.
Blue red		502 518		448	504 517						3
		520 523									
Yellowish red	54 516	512									3
Scarlet	53 64	55									
Bluish scarlet	56										1
Yellow	89 269 329			510	8	94		 		4	
Reddish yellow		84									
Orange yellow		14				13					
Greenish yellow			425			10					
Orange	18 97	17 85						86			
		95									
Blue	439 476	650	692								
	480 655			. 4							
Green blue	440										
Violet	468	452			451						
Blue violet	464										
Grayish violet	287										
Green	433 434	398		435							
Blue green		427									
Yellow green			428								:
Brown	101 137										
D. 135-1 h	139	001		108							
Reddish brown	100	201		197							
Blue black  Blue to bluish red to violet, according to brand	188										
Oil-soluble colors	10	11									
Total	49 60										
	35	20	4	4	5	3	1	1		1	7.

The six colors comprising Class II (those not listed in the Green Tables but whose composition was avowed or disclosed) are one black, one yellow, and one orange among the water-soluble colors, and three yellows among the water-insoluble colors. Therefore two-thirds of the United States market as thus disclosed calls for a total

of 80 different chemical individuals, of which 73 are water-soluble and 7 are water-insoluble and are used as oil or fat colors. The remaining third of the makers or dealers either do not possess the information or are unwilling to give it. No attempt has yet been made to enter systematically into this unknown region, but careful examination warrants the belief that it can add nothing of material value to the data already obtained which show a total of 23 shades for the 73 water-soluble coal-tar colors, summarized as follows:

#### Number of coal-tar colors.

5 red shades	26	3 green shades	6
		2 brown shades	
•		1 blue-black shade.	
8		1 black shade.	
		1 blue to violet shade	
o viole o shades	U	I bide to violet shade	

The water-insoluble colors numbering 7 are not included, but will be treated separately (p. 159).

This view of the state of the United States market at or about the middle of the year 1907 is without question a true reflection of that market as far as it goes and the actual extent of the coal-tar color market beyond those covered by this canvass of it is not likely to be very great. In support of this view is the interchangeable treatment of formerly patented products, the great lapse of time since a new food color was discovered, and the fact that out of the 74 colors submitted and contained in the Green Tables only 23 are now less than 25 years old, and none is less than 16 years old.

All the Green Table numbers and the number of sources out of a possible 12 offering them are given in the following table:

Number of sources, out of a possible 12, offering colors designated in 1907.

Green Table No.	Sources.	Green Table No.	Sources.	Green Table No.	Sources.	Green Table No.	Sources.
1 4 8 9 10 11 13 14 17 18 49 53 54 55 66 60 64 64 85	10 5 1 1 2 6 2 2 1 1 1 1 2 1 2 1 2 2 2 2 1 1 1 2 1 2 1 1 2 1 1 2 1 1 1 1 2 1 2 1 1 1 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	86 89 94 95 97 101 103 104 105 106 107 108 137 139 146 169 189 188	8 1 6 2 1 1 6 1 1 5 7 2 1 1 2 1 1 2 1 1 2 1 1 2 2 1 1 1 2 2 1 1 1 2 2 1 1 1 2 2 1 1 1 2 2 2 1 1 1 2 2 2 1 1 1 2 2 1 1 2 2 2 2 1 2 2 1 2 2 2 2 2 2 2 2 1 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	240 269 287 329 398 425 427 428 433 434 435 439 440 448 451 452 462 464 468	1 1 1 1 2 3 2 3 1 4 1 4 1 4 1 2 2 2 3 1	476 480 502 504 510 512 516 517 518 520 523 584 601 655 667 692	1 1 2 5 4 3 1 5 2 2 2 2 1 1 2 1 2 3

<sup>1</sup> Italicized figures indicate colors permitted by F. I. D. 76.

Nine manufacturers sent 261 specimens, an average of 29 each, distributed as follows: 70; 38; 20; 20; 15; 43; 25; 18, and 12. Two importers sent 5 specimens each, and one 13, a total of 23 specimens, and an average of 8. These figures reflect a diversity of opinion as to what is needful for food coloring, since each one of these 12 makers or importers believed that for all practical food-coloring purposes his selection was complete and sufficient.

Classifying the 284 specimens as red, yellow, brown, orange, blue, green, violet, and black, the following table is obtained showing the different requirements of each of the 12 makers or importers to produce the necessary shades of the eight colors mentioned:

Total specimens submitted, grouped by makers and colors, showing number of shades required by each.

Maker's number.	· Red.	Yellow.	Brown.	Orange.	Blue.	Green.	Vio- let.	Black.	Number of shades wanted by each.	Total speci- mens.
1 2 3 4 5 6 6 7 8 9 10 11 12 12	25 10 7 11 6 4 6 5 6 13 3	18 6 5 10 2 3 2 4 4 10 1	14 2 10 2 1 1 2	4 1 2 5 3 1 2 1 3 3 1	2 3 1 2 4 3 1 1 1	5 1 1 2 2 2 1 1 4 4 4	2 2 1 3 2 1 1 2 1	1	77777755667333	70 25 18 43 20 15 12 13 20 38 5
Total Per cent Maximum Minimum Average Permitted	99 34.86 25 3 8.25 3	23. 24 18 1 5. 50	11. 27 14 2. 75	26 9. 15 5 2. 17 1	7.75 5 1.83	22 7.75 5 1.83	1.25 5.28 3	0.70 1 0.17		284 100

From this table it appears that not one of the 12 sources desired all of the 8 shades into which the 284 specimens are classifiable to make up a complete set of food colors; 7 out of the 12 sources wanted 7 of the 8 shades; 2 sources wanted 6 out of the 8 shades; 1 source wanted 5 of the 8 shades, and 2 sources were content with 3 out of the 8 shades.

It will be noticed that the permitted list given in Food Inspection Decision No. 76 provides for 7 dyes covering 5 out of the 8 shades of the above classification. The 3 missing shades are brown, violet, and black; the shades provided are red, yellow, orange, and blue.

It will also be noticed that on the whole 6 out of the 8 shades were not wanted by one or more of the 12 sources. The italicized shades are the ones not provided for by the permitted list of Food Inspection Decision No. 76.

Brown was not wanted by 5, nor orange by 1, blue by 3, green by 2, violet by 3, nor black by 10.

The combinations not wanted were as follows: Five sources omitted black only; 2, brown only; 1 blue and black; 1 brown and black; 1 green, violet, and black; and 1 brown, blue, green, violet, and black.

Not one of these 12 sources wanted only the three colors not found on the permitted list, and 2 sources did not want any of the three missing shades nor two of the permitted colors.

In view of this large difference of opinion among the 12 sources as to the shades needed to make a complete set of food colors, the 5 shades selected for the permitted list of Food Inspection Decision No. 76 seem reasonably close to any consensus of opinion derivable from the tabulation of the collected facts.

#### II. PURPOSES OF FOOD COLORING.

The use of any color which conceals inferiority, or which gives an article an appearance better than it properly possesses is, of course, illegitimate, and such cases are not here considered. Among such uses may be mentioned that of color in pastry to impart a yellow color thereto, implying the presence of eggs, when they are either wholly absent or are not present in sufficient quantities to produce a shade of color which would indicate a superior quality. Such coloring is frequently resorted to in macaroni, spaghetti, noodles, and the like, and it has also been stated in the literature that such coloring has the additional function of concealing dirt actually present in the flour.

The addition of red coloring matter to meat products to give them an appearance of freshness which they do not of themselves possess; the addition of red coloring matter to strawberry, raspberry, and similar jams, jellies, and preserves, to give them a color indicative of exceptional quality, even though they may contain none of the fruit whose presence is intimated by the label on the product; the injection of red coloring matter into ordinary oranges to give them the appearance of blood oranges; the sprinkling of lemons and oranges with green coloring matter to give them the appearance of a particular origin or of a particular state of ripeness when such origin or state of ripeness is without foundation in fact; the injection of red coloring matter into watermelons to give them the appearance of ripeness, which ripeness they do not possess, are practices met more or less frequently.

Among the purposes for which food colors are said to be used and the foods so colored, the following are mentioned in the literature:

#### In European countries.

<sup>1.</sup> Macaroni is colored with Dinitrocresol (2) (Arch. Pharm., 3d ser., v. 22, p. 621) and Martius Yellow (3) (Weyl, Handbuch).

<sup>2.</sup> Cordials and liqueurs with Dinitrocresol (2) (Arch. Pharm., 3d ser., v. 22, p. 621).

- 3. Oranges: Biebrich Scarlet (163) (Weyl, Handbuch).
- 4. Pastry: Dinitrocresol (2) (Weyl, Handbuch). 5. Butter: Dinitrocresol (2) (Weyl, Handbuch).
- 6. To whiten flour: Anilin blue (457) (Zts. Nahr. Genussm., 1906, v. 12, p. 298).
- 7. Noodles are colored to cover up cigar butts, burnt matches, mineral oil, etc. (Zts. Nahr. Genussm., Vol. II, p. 1018).

#### In the United States.

- 8. Jellies, fruit sirups, soda sirups, jams, ketchup, cheap cordials, lemon extract, milk, butter, cheese, ice cream, confectionery, pastries, flavoring extracts, mustard, cayenne pepper, sausage, noodles, wines, and liqueurs (Winton, Connecticut Agricultural Experiment Station Report, 1901, pp. 179–182).
- 9. Cattle feed is colored yellow (Gudeman, J. Amer. Chem. Soc., 1908, v. 30, p. 1623). 10. "Egg color" (399); "Macaroni color" (94); "Tomato catsup color" (105); "Raspberry color" (103); "Mustard color" and "Pie filling color" (4); "Orange color" (87), and "Strawberry red color" (55) are corresponding United States commercial food color names and their corresponding Green Table numbers (Meyer, J. Amer. Chem. Soc., 1907, v. 29, p. 895).
- Dr. E. Ludwig, of Vienna, stated, upon the authority of Dr. Schacherl, at the International Congress of Medicine held in Budapest, in August, 1909, as follows:

The rather widely distributed practice of coloring baker's goods yellow, such as cakes and the like, further the yellow coloring of pastry, macaroni, noodles, and so forth, has as its function the representation of a very large egg content in them; this coloring has been made very convenient because there are in commerce colors intended specifically for this purpose and designated "egg substitute" and which have nothing whatever in common with egg yolk.

Marmalades such as apricot, raspberry, and currant marmalades are frequently found in a colored condition in commerce; in this case the purpose of the coloring is frequently to cover up adulteration; the adulteration may consist in an admixture of a cheap fruit pulp, particularly apple pulp, or in an addition of glucose sirup. Since these admixtures do not possess the color of the marmalades they are simply helped along by the aid of color.

Old fruit sirups are toned up with color and then sold as fresh sirup.

Red colored fermentation vinegar and red colored vinegar essence as well as vinegar made from such essence are in commerce; such coloring has for its purpose to represent the product as "genuine red wine vinegar," which in some countries is highly desired.

So-called "beer color," said to be an extract of roasted malt (malt caramel), is in fact nothing but ordinary sugar caramel and is frequently from time to time publicly advertised; breweries themselves do not use this preparation, but it has been frequently shown that in small taverns by means of this color local beer was converted into Bavarian beer.

The wholesale coloring of coffee beans serves the purpose of representing a better quality than it actually is.

The coloring of cocoa and chocolate by the use of mineral additions and also of coaltar colors was often proven; in this case the coloring serves exclusively to cover up poor quality. In the case of good products such coloring is not practiced.

Colored sausages, and in fact such with a colored meat body as well as such with a colored casing, are frequently colored; coal-tar colors and cochineal serve this purpose, the latter, however, only for the meat. This coloring is to preserve in old goods the appearance of fresh goods.

The green canned goods of commerce are almost all colored with copper compounds.

Tomato pulp frequently comes into commerce colored with a coal-tar color; the purpose of such coloring is to impart to the goods the appearance of having been prepared with extraordinary care. In all these cases it is not at all a question of a harmless change of the natural condition of the food product, but of improper manipulations which are adapted to deceive the purchaser as to the real value of the goods; even, indeed, to mask the danger to health.

#### III. FOOD-COLOR REQUIREMENTS.

#### ADAPTABILITY FOR SPECIAL PURPOSES.

Not all coal-tar colors are adapted for use in food products. Colors are the more desirable for this purpose the higher their tinctorial power, and the greater the resistance they offer to the action of the materials with which they are to be used, and under the conditions existing. Obviously only such colors as of themselves have their tinctorial properties fully developed can be used, and all such colors as require a mordant to develop or bring out the color are not fit for nor capable of use in food products.

Further, if the colored material is subjected to varying temperatures in the process of manufacturing foods, it should be able to withstand the effects of such temperatures, as, for example, in the manufacture of candies. The colors should also withstand the action of reducing agents, such as are generated in the course of fermentation and decomposition of the food product, or where a preservative such as sulphur dioxid is added to the food product to minimize the effect of decomposition of the food upon the color. Such colors are put on the European market, and perhaps, but not necessarily, on the United States market with preservatives added to them. Most of the coal-tar colors are susceptible to the action of sulphur dioxid, particularly when the latter has been used in the decolorizing of glucose, and Uranin (510) is one of the colors found to have the greatest resistance to the sulphur dioxid which may remain combined in candy.

For example, the book entitled "Henley's Twentieth Century Book of Receipts, Formulas and Processes," published in 1907, on page 359, says of sausage color:

It is absolutely necessary in using aniline colors to add a disinfectant to the dyestuff solution, the object of which is, in case the sausage should commence to decompose, to prevent decomposition of azo-dyestuff by the disengaged hydrogen. Instead of boracic acid, formaline may be used as a disinfectant.

- J. Fraenkel (Arbeit. Kaiserl. Gesundh. 1902, v. 18, pp. 518-521; abst. Zts. Nahr. Genussm., 1902, v. 5, p. 986) reports as follows on the composition of colors used in coloring sausages, meats, and preserves:
- 1. Blood color: Moisture, 15 per cent; common salt, 6.6 per cent; borax, 21 per cent; and Ponceau 2 R (G. T. 55).

- 2. Blood red for meat juices: Liquid of a specific gravity of 1.0163 not affected by acids or alkalis and containing 27 per cent total solids; of these total solids 31 per cent were salt, 12 per cent borax, and the remainder Ponceau 2 R (G. T. 55).
  - 3. Casing red: This powder contained Orange II (G. T. 86).
  - 4. Sausage red: A liquid containing Eosin.
- 5. Lobster color: A liquid of specific gravity 1.0064 containing 1.64 per cent of solids, of which 10.9 per cent were salt and the remainder Ponceau R T (G. T. 44).
- G. Possetto (Zts. Nahr. Unters. Hygiene Waarenk. 1891, v. 5, p. 105) cited the following 15 colors as being used for the coloring of pastry:

G. T. No.	G. T. No.
Martius Yellow 3	Tropæolin 00
Victoria Yellow	Tropæolin 000 No. 1
Naphthol Yellow S 4	Tropæolin 000 No. 2
Aurantia Yellow 6	( 17
Acid Yellow G 8	Chrysoidin
Acid Yellow R 9	$\begin{array}{ c c c c c }\hline \text{Chrysoidin} & & & & & & \\ \hline \text{Chrysoidin} & & & & & \\ \hline & & & & & \\ \hline & & & & & \\ \hline & & & &$
	Azoflavin
Tropæolin 0	
A7 '- CI-CC / F.NT 4 - 1	

Algerian Saffron (a mixture of Nos. 4 and 86 and crocein).

Prussian Saffron (composition not given).

"Blood-red" on the American market is starch colored with red coal-tar colors (*Ibid.*, 1896, v. 10, p. 114).

"Butter yellow" is a clear saponifiable oil of reddish-yellow color, containing 3 per cent of anilin-azo-dimethylanilin (No. 16 of the Green Tables).

#### PROPORTION OF COAL-TAR COLOR USED.

The amount or proportion of coal-tar color used has been variously stated. On page IV of the Leffmann translation of Weyl's book entitled "The Sanitary Relations of the Coal-Tar Colors," it is stated that 1 ounce of Auramin (G. T. 425) will color 2,000 pounds of confectionery, which means 1 part of color in 32,000 parts of colored product.

Frentzel (Zts. Nahr. Genussm., 1901, v. 4, pp. 968-974), on authority not given, says that for sirups 1 part of color is used to from 4,000 to 5,000 parts of sirup; in colored sugars 1 part of coloring matter to from 1,333 to 4,000 parts of sugar; and in flour 1 part of coloring matter to from 666 to 1,000 parts of flour.

In pastry 1 to 100,000 parts (Zts. Nahr. Unters. Hygiene, Waarenk. 1893, v. 7, p. 34).

In chapter VII (p. 47), sections 15, 16, 17, and 18, are brought together statements made before the commission on regulations for the Federal food and drugs act, as to the amount of color contained in colored food products. Briefly these are as follows: Confectionery, 1 part of color in 3,500 parts of product; beverages 1 part of color in 128,000 parts, 256,000 parts, 1,024,000 parts; butter, 420 grains of color to 1,000 pounds of butter; or 1 part of color to 16,666 parts of butter.

One ounce of color to 30 pounds of "colored food;" the colored food was not further defined; which means 1 part of color in 480 parts of colored product.

It has further been represented that 1 part of color is sufficient

to whiten 250,000 parts of yellow sugar.

From time to time others have presented information as to the amount of color used in food products. All of these data available have been tabulated, showing the number of parts of colored product containing 1 part of coal-tar color, arranged in the order of the amounts present:

Food <sup>1</sup>	100	Confectionery	24,576
Do <sup>2</sup>	480	Do	30,000
Flour	666	Do	32,000
Do	1,000	Beverages	80,000
Sugar	1, 333	Pastry	100,000
Confectionery	3,500	Beverages	120,000
Sirups	4,000	Do	128,000
Sugar	4,000	Confectionery	192,000
Sirups	5,000	Whitening sugar	250,000
Confectionery	12,800	Beverages	256,000
Butter	16, 666	Do	1,024,000
Confectionery	20,000		

Grouped according to the kind of material colored, the ranges given are as follows:

Beverages 80,000;	Food <sup>1</sup> 100;
120, 000; 128, 000;	<sup>2</sup> 480
256,000; 1,024,000	Pastry
Butter 16, 666	Sugar
Confectionery	4,000
12,800; 20,000;	Sirups
24, 576; 30, 000;	5,000
32,000; 192,000	Whitening sugar 250, 000
Flour 666;	
1,000	

These statements have emanated from persons presumably acquainted with the facts of their own practice, and if that presumption is correct it appears that there are wide variations in practice not only among individual users, but for individual colors. No attempt has been made to prove or disprove these statements by actual determination of the amount of color contained in commercial colored food products.

The 80 chemical individuals on this market for food-coloring purposes, it can be fairly assumed, have been tested and tried out as to their utility, and in this respect further tests were regarded as superfluous and therefore have not been undertaken.

<sup>1</sup> Kind not definitely stated.

<sup>&</sup>lt;sup>2</sup> Said to be for preserved tomatoes.

## SUITABILITY OF SHADES OF PERMITTED COLORS AND MIXTURES OF SAME.

The shades produced by the seven permitted colors are, respectively, yellow, orange, blue, green, red, bluish scarlet, and brilliant cherry red. As statements are found in the literature against the use of all of the chemical individuals producing a brown or a violet shade, it will be necessary to produce these shades by a proper combination of two or more of the permitted colors. So far no criticism with regard to the shades produced by the seven colors themselves and by their appropriate mixture has been made that has been substantiated.

Objection has been made to the violet producible from blue and red, on the ground that when applied to a food product, such as candy, the component parts do not evenly fix themselves upon the material. This objection, however, has not been pressed and probably is not well taken, because of the fact, frequently reported, that few, if any, coal-tar colors are used without admixture of one or more other colors to shade or to tone the original color. This criticism, therefore, of the use of mixed colors can be regarded as not a serious objection.

One criticism urged with considerable persistency against the seven permitted colors was that none of them would withstand the action of the organic acids ordinarily found in beverages such as lemonade, and it was suggested that no color was proper for use for such purposes which would not withstand, unaltered, for a period of 12 hours the action of a 10 per cent solution of citric acid. In urging this objection substitutes were suggested for the permitted colors. The substitutes so urged were Tartrazin (94), Azorubin (103), Orange II (86), Ponceau 4 GB (13), and one other color designated as Scarlet SR, of whose chemical composition no information whatever was forthcoming. (The numbers in parentheses refer to the Green Tables.) Of the five colors suggested it can be said that concerning all but No. 103 adverse statements are found in the literature, and No. 86 is specifically regarded by every observer but one as being thoroughly poisonous. The suggested substitute list is, therefore, objectionable on the ground of injuriousness to health.

To test the validity of the assertion that none of the permitted colors could withstand the action of citric acid, solutions of the suggested colors, as well as of the permitted colors, each one in a thousand, were submitted to the action of citric acid, added in such quantity that it amounted to 10 per cent of the total bulk of solution. This experiment showed that Tartrazin is reddened by citric acid, whereas Naphthol Yellow S loses in tinctorial power to a slight extent. The shade produced by Ponceau 4GB can be closely imitated by a mixture of Naphthol Yellow S, Orange I, and Amaranth, all per-

mitted colors. There is no choice whatever in the shade produced by the desired Azorubin and the permitted Amaranth, nor is there any difference in behavior toward citric acid. The difference in the shade between the desired Orange II and the permitted Orange I is so small that it requires a side-by-side comparison to distinguish between them. Moreover, the desired Orange II produces a precipitate when brought in contact with the citric acid, whereas the permitted Orange I does not so precipitate. The permitted Erythrosin is, of course, completely precipitated by the citric acid. The permitted Light Green and Indigo disulphoacid are weakened in tinctorial power by the addition of the citric acid. Of these colors the only ones used to any extent in beverages, so far as either the suggested or permitted list is concerned, are red, yellow, green, and orange.

As has been shown the permitted reds equal the desired reds and the permitted orange is better than the desired orange. The tinctorial power of the permitted yellow is not so great as the tinctorial power of the desired yellow, but this difference is so slight that the objection urged against the list of permitted colors, namely that they were so poor in quality that they had destroyed a profitable and lucrative business in the coloring of beverages, is untenable in view of the fact that, assuming a price of \$1 per pound for Tartrazin, and 40 cents for Naphthol Yellow S, and using them in the proportions necessary to produce a lemonade color in a 10 per cent citric acid solution, it would take 5,000 quarts of finished lemonade to cause an increase of 1 cent in the cost of the production of the colored food product; that is, it increases the price per quart by one five-thousandth of a cent.

It has also been urged that the permitted green is not good enough for cordials and liqueurs, and that it is impossible to bring about the proper green by the use of the permitted yellow and blue. This criticism, however, has not been persisted in; the fact is that mixtures of the permitted yellow and blue can be made so as to obtain any desired shade of green, having a yellow or blue cast, and great clarity and brilliancy. How these mixed colors would look after a long period of time has not been ascertained.

It has also been said that the permitted red, Amaranth, is not a color suitable for the coloring of strawberry jams and it has been urged that the same chemical individual under another commercial name is better than the permitted red. This criticism has not been pressed, probably for the reason that it can not be substantiated.

Again it was claimed that the deposits to be noticed in bottled lemonades were due to Naphthol Yellow S, but solutions of Naphthol Yellow S in citric acid have remained without deposit for upward of 15 months; it is possible that such precipitation, if observed, may be due to an admixture of the nonpermitted Orange II with Naphthol

Yellow S, and this difficulty can be obviated by the use of the permitted Orange I, which does not precipitate in the citric acid, as shown.

Naphthol Yellow S has been objected to on account of the bitter taste it is said to impart to the beverages to which it is added, and steps have finally been taken by those interested to have another yellow placed on the permitted list.

It has also been objected that the permitted blue is not suitable for the coloring of sugar, first, because it is soluble, and second, because of the unsatisfactory shade. It may be sufficient, in answer to this criticism, to state that there was no insoluble blue offered on this market, and there was no blue other than the one permitted offered against which adverse statements did not exist in the literature, and in view of this state of affairs the criticism may be said to be not well taken.

As against all these specific criticisms it has been repeatedly stated by those in a position to know that they have found no difficulty whatever, by suitable mixtures of permitted colors, in reproducing any desired shade of any desired quality, not even excepting browns and violets.

Considering all of these criticisms, therefore, the conclusion seems reasonable that there is no serious or permanent objection to be made against the seven colors selected, either as to qualities for food-coloring purposes or range of producible shades.

## IV. CONFORMITY OF FOOD-COLOR MARKET, 1907, TO RECOM-MENDATIONS OF THE NATIONAL CONFECTIONERS' ASSOCIA-TION, 1899.<sup>1</sup>

Having thus shown that the food-color market of the United States contains not less than 80 coal-tar colors which are distinct chemical individuals, of which 74 are entered in the Green Tables and 6 are not, the next question to be considered is whether all of these substances are harmless and fit for use in food products.

As a guide in determining this point the "Official circular from the executive committee of the National Confectioners' Association of the United States," pertaining to colors in confectionery, dated February 1, 1899, may well be considered.

The function of this circular is said to be "to throw light upon the vexed question of what colors may be safely used in confectionery," evidently because "there may at times be a doubt in the mind of the honest confectioner as to which colors, flavors, or ingredients he may safely use and which he may reject."

The circular also states that "but infinitesimal amounts of color (coal-tar colors) need be or can be used to give the desired effects,"

and in view of this statement as to quantity it must be self-evident that a color harmful when used in the small quantities said to be used in confectionery is certainly harmful when used in the large quantities used in coloring other food products. This circular under the heading, "Colors that are injurious and therefore to be rejected—Harmful organic colors," enumerates 21 coal-tar colors. Of these 21 colors, 13, or 61.9 per cent, were among those submitted, and whose composition was stated by reference to Green Table numbers; the Green Table numbers of these colors, together with the number of sources from which they were obtained, follow:

Green Table numbers.	Number of sources.	Green Table numbers.	Number of sources.
17	2	197	
84	2	398	2
86	8	584	1
95	2	650	27
106	5		

Out of these 13 colors 3 each came from one source; 7 each came from two sources; 1 came from four sources; 1 came from five sources, and 1 came from eight sources, out of a possible 12; that is, one was wanted by more than half the sources.

It is further to be noted that of the trade names given to the submitted products of Class I the following appeared among the harmful list of this circular and also were found in identical form and spelling on the labels of the submitted products:

- 1. Bismarck Brown.
- 2. Chrysoidin R.
- 3. Chrysoidin Y.
- Mandarin G extra.
   Naphthol Green B.
- 6. Napthol Yellow.
- 7. New Coccine.

- 8. Orange A.
- 9. Orange A extra.
- 10. Orange G.
- 11. Orange II.
- 12. Scarlet.
- 13. Vesuvin B.
- 14. Crocein Scarlet 5 B.

The following parallel will serve to show the great resemblance between the names given to the harmful colors of the circular and those found on the samples submitted:

Labels of submitted sample
Methylene Blue B.
Methylene Blue D.
Methylene Blue O.
·
New Coccine O Z.
New Coccine Z.
Naphthol Green.

Circular's	: harm	ful	list.
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### Labels of submitted samples.

Orange II	
Victoria Yellow	Victoria Yellow Conc. Z.
	Victoria Yellow Conc. T Z.
Acid Yellow	Acid Yellow G.
Bismarck Brown G	Bismarck Brown B.
Bismarck Brown T	
	Bismarck Brown Dark
	Bismarck Brown B. X.
	Bismarck Brown T D.
	Bismarck Brown Y Bril.
	Bismarck Brown Y Dark.
	Bismarck Brown 2 R X.
Chrysoin	
Om y both	Chrysoin G E Z.
	Chrysoin R E Z.
	Chrysoin R Z.
Cochineal Red A	
Crocein Scarlet 3 B.	
Crocein Scarlet 7 B	Crocein Scarlet 10 B.
Crocein Scarlet 7 B	
Fast Brown G.	Fast Brown N.
rast Drown G	
E4 V-11	Fast Brown O.
Fast Yellow	
	Fast Yellow O 3 3.
	Fast Yellow O 3 4.
	Fast Yellow Y.
Imperial Scarlet, in powder, extra	
Safranin	Safranin S P.
Safranin A G extra.	
Safranin AGT extra.	
Safranin Conc.	
Safranin extra G.	
Safranin FF extra No. 0.	
Safranin G extra GGS.	
Safranin G 000.	
Safranin T.	

This comparison disclosed a considerable lack of conformity between the United States food-color trade in 1907 and the circular of February, 1899, upon whose preparation for seven months prior to its date "a great deal of thought and labor have been given to a thorough investigation of the whole subject of colors in confectionery," in which the committee has been largely aided by the researches of the association's chemist and by the results of his analytical tests" and whose "classifications have been carefully made, and are based upon the authority of the eminent chemists, Prof. Koenig and Prof. Weyl, upon the resolutions of the Swiss chemists and upon the French ordinances regarding the coloring of food products," and which list was expected to be "of value to color

dealers and chemists," and also was published to assist the confectioner in obeying "the letter and the spirit" of the pure-candy laws.

Under the heading "Colors that have been shown to be harmless as used in the confectioner's art, harmless organic colors," this circular enumerates 36 colors, for 4 of which there are no Green Table numbers. Of the 32 colors having Green Table numbers, 20, or 62.5 per cent, were among those colors submitted. The Green Table numbers of these colors, together with the number of sources from which they were obtained, follow:

Green Table numbers.	Number of sources.	Green Table numbers.	Number of sources.
4	10	240	
8	5	269	1
9	1	287	1
13	6	427	2
55	5	448	4
65	2	451	5
85	2	462	2
103	6	512	3
105		517	5
107	7	520	2

Of these 20 colors 5 each came from 1 source; 5 each came from 2 sources; 1 came from 3 sources; 1 came from 4 sources; 4 each came from 5 sources; 2 each came from 6 sources; 1 came from 7 sources, and 1 came from 10 sources; that is, only 2 were wanted by more than half the makers or importers, and only 4 by half the sources.

It will be noted that this circular provides for a total of only 57 different coal-tar colors; the number of avowed colors submitted reached 80, or 23 in excess of this number, and further, that out of the 57 colors referred to in this circular only 33 appeared among those colors submitted whose composition was acknowledged, so that for 47, or 58.8 per cent of the avowed submitted colors, this circular is no specific guide. From the data obtainable from this circular, the following tabulation can be made:

Harmfulness of submitted colors based on list in circular.

Data.	Total on market.	Disclosed colors on United States market.	Circular's corre- sponding list.
Harmful Harmless	13 20	Per cent. 16.3 25.4	Per cent. 61. 9 62. 5

These figures disclose a considerable and self-evident disregard of the request, then eight years old, as made by the National Confectioners'

Association of the United States, that coal-tar colors designated by it as "colors that are injurious and therefore to be rejected, harmful organic colors" be not used in confectionery, and by implication that they should not be used in other food products.

In view of this disregard with respect to 13 coal-tar colors out of 33 on the United States market in the summer of 1907, dealt with in this confectioners' list, the conclusion seemed justified that the action regarding other coal-tar colors on the United States market in the summer of 1907 was equally heedless or indifferent, a conclusion which is borne out by the material brought together under section IX.

The necessity of adhering to some unequivocal terminology, as has been done in these pages by referring to the serial numbers in the Green Tables, appears from the following:

Trade names are not definite with respect to the composition of the article sold under a given name, although tinctorially the colors may be substantially equivalent. In the Green Tables there are not less than 29 instances where the same trade name is applied to two or more different chemical individuals. In some cases these chemical individuals are fairly closely related, in others they are only remotely related. When the differences are greater than the presence or absence of a sulpho group or the use of methyl for ethyl or the reverse, the instances are given below:

1.	Cotton Yellow	128	Primulin-azo-m-phenylene-diamin-disulphonic acid
		191	Diphenylurea-disazo-bi-salicylic acid.
2.	Methyl Eosin	513	Methylated tetrabromo-fluorescein.
		375	Dinitro dibromo fluorescein.
3.	New Yellow	88	p-sulphanilic acid azo-diphenylamin.
		91	Nitration product of diphenylamin yellow.
4.	Orange III	23	Meta-nitranilin azo-R salt.
		87	p-sulphanilic acid azo-dimethylanilin.
5.	Orange N	43	Toluidin azo-Schaffer acid.
		88	p-sulphanilic acid azo-diphenylamin.
6.	Orange R	97	o-toluidin-monosulphonic acid azo-betanaphthol.
		15	Anilin-azo-R salt.
		99	Xylidin sulphoacid-azo-betanaphthol.
7.	Toluylene Red	261	Dichloro-benzidin disazo R salt.
		580	Dimethyl diamido toluphenazin.

## The following quotation also bears on this point:

In attempts to group the aniline colors a kind of uncertainty appears even among color chemists. The same trade name does not always correspond to the same preparation. Many preparations are not chemical individuals, but mixtures of related colors. Many preparations are "standardized" for the trade; for example, with dextrin. On account of the patent laws, factory secrets surround the production of many coloring matters, and frequently statements are met with which are directly intended for the purpose of misleading competition. (Hueppe, Die Methoden der Bakterienforschung, 5th ed., 1891, p. 105.)

# V. SOME LEGAL ENACTMENTS RELATIVE TO THE USE OF COAL-TAR DYES.

### LIST OF THIRTEEN FOREIGN LEGAL ENACTMENTS.

The 13 foreign legal enactments compared are as follows:

- 1. The Austrian regulation of March 1, 1886, which forbids No. 483 of the Green Tables and all anilin colors.
- 2. The law of Austria of May 1, 1886, which forbids No. 1 of the Green Tables and all anilin colors.
- 3. The Austrian regulation of September 19, 1895, in which certain colors only are permitted, and all others are forbidden. There are 16 titles of permitted colors in this law, but these are in some cases so elastic and so indefinite as to include 47 entries in the Green Tables.
- 4. The Austrian law of January 22, 1896, in which there are 17 titles, but these are sufficiently elastic to allow of 22 entries in the Green Tables being included in them.
- 5. The German law of July 5, 1887, which specifically prohibits only Nos. 1 and 483 of the Green Tables. The interpretation which seems to be generally placed upon this law is that all other Green Table members are permitted in Germany for use in foods.
- 6. The Italian law of February 7, 1892, forbidding all colors except 9 different titles, which, however, were elastic enough to include 32 entries in the Green Tables.
- 7. The law of Italy of February 7, 1902, which prohibits 37 entries in the Green Tables, and permits 11 specifically.
- 8. The Italian decree of June 29, 1893, in which there were 7 titles of permitted colors sufficiently elastic to include 34 different individuals.
- 9. The Italian decree of March 24, 1895, forbidding four titles covering only four entries in the Green Tables.
- 10. The French police ordinance of May 21, 1885, in which 489 entries in the Green Tables were prohibited.
- 11. The French police ordinance of December 31, 1890, in which 469 entries of the Green Tables were prohibited, and which also permitted under 9 titles 23 entries in the Green Tables.
- 12. The Belgian law of 1891, which specifically forbids only four entries in the Green Tables.
- 13. The law of the Canton of Tessin, dated May 18, 1897, which forbids only 4 specific entries in the Green Tables.

### SUMMARY OF COLORS PERMITTED BY THESE LEGAL ENACTMENTS.

An examination of the 13 legal enactments made in Europe with respect to the use of coal-tar colors in food products discloses considerable difference of opinion as to the harmfulness or the harmlessness of even the same chemical individuals. To prepare an approximate and comprehensive summary of the effect of such legislation, the following plan has been followed:

The provisions of a selected number of laws and regulations, 13 in number and dating from 1883 to 1902, were read with respect to permission or prohibition of the 695 chemical individuals listed in the Green Tables. The effect of each law upon each separate entry in the Green Table numbers was noted, either as permitted, forbidden, or noncommittal when the law was silent upon such entry; the laws were read with the understanding that what was not forbidden was permitted, and what was neither forbidden nor permitted was noncommittal; if this were an incorrect or improper procedure the number of permitted colors would be reduced but not augmented. For the purposes of this first approximation, no attempt was made to segregate those specifically permitted from those permitted by blanket expressions or phrases.

For the purposes of a side-by-side comparison, the term "index number" was coined; this "index number" gives in the first place the number of legal enactments that permit the color; in the second place the number of enactments that forbid it; and in the third place the number of enactments that are silent or noncommittal. Thus: 283 as an index number would mean 2 enactments permit, 8 forbid, and 3 are noncommittal; so that an index number with the highest hundreds would have the greatest number of permissions, and with the highest tens would have the highest number of prohibitions. With this understanding of these terms, the following table of so-called index or "P. F. N." numbers is offered:

Thirteen legal enactments classified by Green Table numbers and the "P. F. N. figure," or "index number."

Total number of Green Table entries.	Index number.	Green Table numbers.
1	184	483.
i	274	534.
1 1 3	2, 10, 1	1-3.
217	364	394-411, 415-426, 429-434, 4351, 436-445, 484-492, 528-561, 564-583, 585-598, 600,602-649,651-691,693-695.
1	373	103.
366	382	6,7,10,11,12,14,16,19-22,35-40,42,43,45-54,58-64,66-83,89,90,93,94,96-101,104-106,109-113,115-145,149,151-156,158,159,161,162,164-168,
	1	172, 197, 199-243, 245-393, 412-414, 493-511, 524-527, 650.
29	391	446, 447, 449, 450, 453, 455, 456, 458–461, 463–476, 478, 479, 481, 482.
3 1	454	428, 563, 599.
1	463	692.
22	472	5, 8, 9, 23–34, 84, 86–88, 91, 520, 523.
7	481	95, 452, 454, 477, 513, 519, 522.
2	544	562,601.
2	562 571	515, 516. 480, 514, 521.
7 2 2 3 3	643	44, 114, 427.
11	652	13, 15, 85, 92, 108, 146–148, 150, 160, 517.
	661	17, 18, 41, 169, 512, 518.
2	670	451, 462.
6 2 6 6 1 2	742	4, 55, 56, 57, 102, 244.
6	751	65, 107, 163, 170, 198, 448.
1	850	457.
2	931	157, 171.
2	931	157,171.

<sup>1</sup>Italicized figures indicate colors permitted by F. I. D. 76.

From the table it appears that there is no one Green Table number that is permitted by each and all of these 13 legal enactments, nor is there any one color that is prohibited by each and all of these 13 legal enactments; and as late as 1902 there was considerable confusion as to what should or should not be permitted or forbidden. That the European enactments were not in reality consistent or effective appears from the following:

1. In respect to the use of coal-tar colors, the views as to their harmfulness or harmlessness are very divergent, and this uncertainty is expressed in the various legislative enactments. (v. Raumer, Zts. Nahr. Unters. Hygiene & Waarenk., 1895, v. 9, p. 207.)

2. After eating groats, which no doubt were free from ordinary poisons but had been colored with Martius Yellow, a whole family became sick. Since this coloring matter is not mentioned among those which, under the law of July 5, 1887, are forbidden for the purpose of coloring articles of food, a complaint could not be lodged. Nevertheless, the use of Martius Yellow for the coloring of articles of food would seem to be dangerous, since this coloring matter exerts poisonous effects. It is a weakness in the law that coal-tar colors, of which new members are continuously appearing on the market, and whose physiological action is unknown, should be at all permitted for the coloring of articles of food. (Dietrich, Th., Jahresberichte der landwirthschaftlichen Versuchsstation, Marburg, 1900–1901, p. 13; abst. Zts. Nahr. Genussm., 1902, v. 5, p. 364.)

On account of their large number and the great diversity of opinion as to harmfulness of some coal-tar colors and the harmlessness of others therein reflected, no attempt was made to collect all the legal enactments and regulations made with respect to coal-tar dyes. The foregoing are typical of the remainder.

## COLORS SAID TO BE PERMITTED UNDER THE GERMAN LAW OF 1887.

That some of the laws did not employ specific terms but used those possessed of a great degree of elasticity appears from the following taken from Weyl's "Sanitary relations of the coal-tar colors," page 38, concerning the Chamber of Commerce and Trade of Sonneberg which declared on December 4, 1887, that the German law of July 5, 1887, allowed the unrestricted use of—

All blue and violet anilin (that is coal-tar) colors, all ponceaus, all orange colors, Methyl Green, Brilliant Green, Malachite Green, Chrysoidin, Naphthol Yellow, Martius Yellow, Eosin, Phloxin, Safranin, Erythrosin, Fuchsin, Phenylene Brown, and Anilin Black.

This amounts to not less than 233 permitted colors, as will now be shown.

Using the Green Tables as a guide it will be found that under the above ruling there are to-day no less than 107 blue coal-tar colors which could be used for food coloring. They are arranged as follows, showing the comments on same in the literature.

### BLUE COLORS.

Unfavorable: 478, 479, 488, 490, 572, 602, 639. (Total, 7.)

Favorable: 477, 599, 600, 692. (Total, 4.)

Contradictory: 287, 457, 480, 563, 601, 650, 689. (Total, 7.)

Not reported on: 36, 83, 119, 142, 175, 189, 209, 246, 247, 254, 257, 263, 266, 288, 290, 291, 292, 293, 294, 295, 301, 302, 310, 311, 314, 315, 316, 317, 318, 319, 323, 327, 345, 347, 348, 351, 352, 356, 359, 364, 430, 432, 439, 440, 442, 444, 456, 473, 476, 481, 482, 487, 492, 509, 542, 551, 556, 558, 559, 560, 562, 569, 595, 598, 608, 611, 612, 615, 618, 619, 621, 625, 627, 628, 634, 637, 638, 640, 641, 642, 643, 652, 655, 656, 657, 664, 682, 693, 694. (Total, 89.)

Similarly there would be 50 violets which are classified as follows:

#### VIOLET COLORS.

Unfavorable: 620, 649. (Total, 2.) Favorable: 467, 593. (Total, 2.)

Contradictory: 450, 451. (Total, 2.)

Not reported on: 30, 34, 36, 118, 176, 179, 207, 244, 246, 248, 252, 256, 273, 320, 336, 338, 340, 342, 343, 444, 452, 454, 455, 463, 464, 465, 466, 468, 469, 470, 471, 472, 474, 486, 506, 507, 525, 552, 579, 581, 585, 592, 613, 625. (Total, 44.)

Similarly there would be 18 ponceaus which are classified as follows:

#### PONCEAUS.

Unfavorable: None.

Favorable: 169, 448. (Total, 2.)

Contradictory: 13, 15, 55, 160, 163. (Total, 5.)

Not reported on: 44, 56<sup>1</sup>, 57<sup>1</sup>, 108, 113, 114, 146, 147, 148, 150, 165. (Total, 11.)

Similarly there would be 35 Oranges which are classified as follows:

### ORANGES.

Unfavorable: 2, 97. (Total, 2.)

Favorable: 85. (Total, 1.)

Contradictory: 14, 18, 43, 86, 87, 88, 95. (Total, 7.)

Not reported on: 10, 23, 47, 54, 99, 100, 136, 162, 196, 217, 218, 222, 225, 235, 236, 265, 275, 392, 406, 408, 409, 529, 531, 545, 547. (Total, 25.)

The remaining 23 colors named are classified as follows:

### MISCELLANEOUS.

Unfavorable: Martius Yellow (3).2 (Total, 1.)

Favorable: Naphthol Yellow S. (4); Eosin (512, 517, 521); Phloxin (521); Erythrosin (517). (Total, 6.)

Contradictory: Brilliant Green (428); Malachite Green (427); Chrysoidin (17, 18, 41); Safranin (584); Fuchsin (448); Phenylene Brown (197). (Total, 8.)

Not reported on: Methyl Green (460, 461); Eosin (514, 515); Phloxin (518); Safranin (583, 585); Erythrosin (516); Anilin Black (577). (Total, 9.)

It will be noticed that names such as Eosin, Erythrosin, and Phloxin appear in more than one classification; thus some of each are favorably reported on and the others are not reported on at all.

<sup>&</sup>lt;sup>1</sup> Nos. 56 and 57 are included among the permitted colors of the Austrian law.

<sup>&</sup>lt;sup>2</sup> The numbers in parentheses following the names are the Green Table numbers.

The 233 colors said by the Sonneberg Chamber of Commerce and Trade to be entitled to unrestricted use in food coloring under the German law of 1887 referred to may also be classified as follows:

Summary of classification of colors permitted under the German law of 1887 according to comments in the literature.

		Comments in the literature.			
· Class.	Total.	Unfa- vorable.	Favor- able.	Contra- dictory.	Not reported on.
Blues. Violets. Ponceaus Oranges Miscellaneous.	35 23	7 2 0 2 1	4 2 2 1 5	7 2 5 7 8	89 44 11 25 9
Total	233	12	14	29	178

Out of these 233 colors only 55, or about one-fourth, have been reported on in the literature and the remaining three-fourths have not been examined at all. To the 55 examined and reported on there may be added 2, namely, Nos. 56 and 57, since they are included among the colors once permitted by law in Austria, thus making a total of 57 examined out of 232. Adding these two to the 14 favorably reported on makes a total of 16. It finally appears that 41 out of 57 colors examined would in the light of present knowledge be improper to be used in food and 12 at least should not be used for such purposes at all.

The classification into unfavorable, favorable, contradictory, and not reported on is based upon the tabulation on page 63; if that is substantially correct the above conclusions are also true.

The foregoing side-by-side comparison of 13 legal enactments, while it makes no claim to being absolutely and wholly accurate in all the classifications or conclusions drawn, is no doubt a fair reflection of the condition of mind of those framing the enactments, and consequently of information upon which those enactments were based; and the conclusion would therefore seem to be justified that the fact that a color has been permitted or has been forbidden by any one or the majority of these legal enactments ought not to constitute a clean bill of health, nor an indictment, as the case may be.

This side-by-side comparison must not be pushed to extremes; indeed the extent to which it can be employed is naturally very limited, and the purpose for which it was made was to reflect in a manner easily and comprehensively grasped the confusion and inconsistencies which very persistently force themselves upon the mind of a person reading those enactments and having in mind the chemical individuals at which they are aimed.

The definite lesson to be learned from this side-by-side comparison is that these enactments in many cases employed terms so vague and indefinite as to permit the use of some bad colors as well as all good ones, that is they were not sufficiently definite to exclude all that were harmful.

# DEFINITENESS AND DETAIL NECESSARY TO EFFECT QUALITY CONTROL.

This apparent state of confusion in legal enactments that preceded the summer of 1907 was a very strong factor in the formation of the conclusion that in order to be effective any law or regulation dealing with coal-tar colors for use in foods must prohibit every coaltar color except certain definite specific ones.

The Austrian laws of September 19, 1895, and of January 22, 1896, provided for quality control by public and other laboratories of the coal-tar colors put upon the market for use in foods; the results of such control, as reported in the Zeitschrift für Nahrungsmittel-Untersuchung, Hygiene und Waarenkunde, 1896, v. 10, p. 335, are as follows:

Coloring matters of commerce are mostly mixtures of various coloring matters, a right which manufacturers will not part with; and further, while it is indeed possible to test the coloring matter in substance, it is nevertheless impossible to test it in the very small amounts which are used in the coloring of foodstuffs and to determine with certainty the identity of the color as to whether it is or not one of the permitted colors.

Of 21 samples of coloring matter examined, 14 were objectionable, partly because of false labeling, or because they were mixtures, partly because they contained poisonous metals, or a forbidden coloring matter. Thus, a so-called "Evergreen" was Naphthol Green B, a poisonous nitroso color; Malachite Green contained zinc; an Acid Magenta and a Rosalin contained traces of copper; Ponceau, Eosin, Brown, and Roccellin contained traces of tin; Orange I and Waterblue contained traces of tin and zinc. The last-named coloring matters were therefore not prepared in proper state of purity.

The authorities in Vienna examined four and rejected two colors. (*Ibid.*, 1898,

p. 107.)

The Swiss authorities exercised control over colors, after they reached the market, with the result that the authorities in Basle examined ten colors and rejected one. (*Ibid.*, 1897, p. 292.)

These facts, together with the knowledge derived by even the most superficial ocular examination of the 294 specimens received in the summer of 1907, played a very great part in the formation of the conclusion that control of quality, in order to be even reasonably effective, must be thoroughgoing, and that colors must be excluded from the market until they prove themselves to be clean, rather than permitted promiscuously and then driven out of the market by the authorities if unsuitable.

The effective quality control of food colors requires careful and searching examination of a kind which can not usually be obtained

by the general purchasing public. The quality of the food colors offered in the summer of 1907 varied greatly, and the substances contaminating them were of such indefinite and probably variable composition (of whose physiological action nothing definite was known and whose quick and certain detection in the colored food product would be very complicated, if not impossible) that quality control of greater efficiency than that exercised by those selling food colors in the summer of 1907 seemed necessary on the part of the authorities having charge of the enforcement of the food and drugs act.

The points of original entry of food colors into the United States food-color market are relatively few, whereas the points of distribution of food colors are very many, the former being less than 20 and probably fewer than 10, while the latter may number up into the hundreds; therefore, not only is the labor and the expense of quality control of food colors reduced to its probable minimum by keeping food colors off the market until they have shown their right to be so used, but also the certainty and the efficiency of quality control

is increased to its probable maximum.

The quality control thus suggested is similar to that exercised by the States of New York, Michigan, and Ohio over salt before it enters the market for human consumption. The method of color control here suggested differs only in degree, not in kind, from the quality control exercised over salt by the States named. Experience has shown such quality control of food colors to be not only practicable but capable of realization without any hardship and but little, if any, inconvenience to those concerned.

### STATE LAWS PROHIBITING THE USE OF COLORS IN CERTAIN FOODS, 1909.

The laws of the individual States of the United States have also restricted the use of coal-tar coloring matters in foods. These restrictions are directed principally against the use of color to conceal inferiority, which restriction is found in almost all the States.

The sale of poisonous coloring matters for foods is prohibited in the State of New York, and in New York and North Carolina the addition

of injurious colors to foods is prohibited.

Minnesota and North Carolina prohibit coal-tar dyes in all foods. Foods and beverages are considered adulterated in North Dakota and Wyoming if they contain aniline dyes or other coal-tar dyes.

Artificial coloring is prohibited in sausages by Colorado and Wisconsin.

Artificial coloring, including, of course, coal-tar colors, must not be added to vinegar in the States of Arkansas, California, Connecticut, Iowa, Minnesota, Missouri, New Jersey, New York, Pennsylvania, Tennessee, Wisconsin, and Wyoming.

Distilled vinegar must not contain artificial color in Ohio and Oklahoma, and must be free from harmful artificial coloring matter in Utah.

In South Dakota oleomargarine must not be colored.

Artificial coloring is prohibited in milk by California, Oklahoma, Pennsylvania, Utah, and Wisconsin and in cream by California Connecticut, Pennsylvania, Utah, and Wisconsin.

Coal-tar dyes are inhibited in cakes, crackers, candy, ice cream, and like products by Virginia. Ice cream is considered adulterated in Michigan if it contains harmful colors.

Forty-six States prohibit the use of poisonous colors in candy. They are as follows: Alabama, Arkansas, California, Colorado, Connecticut, Delaware, District of Columbia, Florida, Georgia, Idaho, Illinois, Indiana, Iowa, Kansas, Kentucky, Louisiana, Maine, Maryland, Michigan, Minnesota, Missouri, Montana, Nebraska, Nevada, New Hampshire, New Jersey, New York, North Carolina, North Dakota, Ohio, Oklahoma, Oregon, Pennsylvania, Philippine Islands, Porto Rico, Rhode Island, South Carolina, South Dakota, Tennessee, Texas, Utah, Vermont, Virginia, Washington, Wisconsin, and Wyoming.

# VI. RECOMMENDATIONS BY ASSOCIATIONS AND INDIVIDUALS AS TO USE OF COAL-TAR DYES AS FOOD COLORS.

### CAZENEUVE AND LÉPINE.

Cazeneuve and Lépine (Bull. de l'acad. de médicine, April 27, 1886, p. 643) says:

We have arrived at the following conclusions:

1. The nitro derivatives are especially poisonous (dinitronaphthol being comparable with picric acid), but the sulphonated product is harmless.

2. Safranin and Methylene Blue are harmful, producing gastric intestinal dis-

turbances, being violent poisons.

3. The following coloring matters are tolerated by man, whether well or affected with Bright's disease; similarly, too, animals (dogs, guinea pigs) without any noticeable disturbances and at rather high doses:

	Probable Green Table Nos.		Probable Green Table Nos.
1. Fast Yellow	9	5. Ponceau R	55
2. Roccellin	102	6. Orange I	85
3. Bordeaux B	65	7. Fuchsin S	462
4. Purple	106 or 107		

Among the nontoxic sulphonated colors we have been able to make out the following list or classification, based upon their power of producing disturbances, proceeding from the least inert to the most inert:

mont one ready mere to the most	d allect c.	*	
	Probable Green Table Nos.		Probable Green Table Nos.
1. Orange I	85	5. Yellow NS	4
2. Bordeaux B	65	6. Fast Yellow	9
3. Ponceau R	55	7. Purple	106 or 107
4. Roccellin	102		

The results of our experiments have led us to the following conclusions:

1. The relative nonpoisonous nature of the azo colors used for coloring wines explains why this artificial coloring has not caused any real epidemic.

- 2. This artificial coloring of wines by coal-tar colors is dangerous. It opens the door to the employment of coloring matters of very variable and noxious properties. Thus Martius Yellow, which is poisonous, has been used for 10 years past to color pastry (3 grams per 100 kilograms) and it may be used to-morrow, perhaps, to color wines mixed with a red or a blue.
- 3. A rigid law against the artificial coloring of wines ought to be promulgated, particularly if this coloration covers detestable practices most prejudicial to the public health. The addition of salicylic acid, glycerin, and tartaric acid, or the acidifying by sulphuric acid, is cloaked by the use of the coloring matter.

Some sulphonated azo-coloring matters are sufficiently inert to enable their being employed as artificial color in foods, bonbons, and liquors. These colors are manufactured according to simple processes which give theoretical yields and no metallic salt, such as mercury, tin, or arsenic participates; sulphate of soda is the only impurity.

In view of the great extent of the use of these coloring matters, it is better to regulate their consumption by tolerating certain of these products rather than to interpose an illusory barrier to their use. Where you can not arrest a stream you can at least regulate its course.

It would be better definitely to classify these substances with respect to their noxious properties, tolerate some and prohibit the others, rather than to be exposed to the consequences of permitting manufacturers to introduce into food, without any scientific control whatever, any products whatever.

These coloring matters should be sold in commerce under the names of harmless colors as determined by analysis. By chemical analysis it would be recognized as to whether we were dealing with one color or with a mixture of two or three colors.

The colors most used are made up of red, yellow, and blue, which apparently imitate the appearance of the wines of the Midi. Thus we have recognized such coloring matters in Roccellin, Naphthol Yellow, and Methylene Blue.

Sulphonated Fuchsin is very much used, combined with a yellow and a blue. This mixture turns green with ammonia, like the coloring matter of wine. In fact, Acid Fuchsin is decolorized by ammonia. The yellow and blue remain intact, and give a green which suggests true wine color.

### SOCIETY OF SWISS ANALYTICAL CHEMISTS.

In 1891 the Society of Swiss Analytical Chemists recommended that certain coloring matters which are to be regarded as harmful to health should not be permitted to be used in the preparation of articles of food intended for sale in which artificial coloring is at all permitted.

The coal-tar colors thus prohibited are identified in the following by their Green Table numbers, only one trade name being given: Picric Acid (1); Dinitrocresol (2); Martius Yellow (3); Aurantia (6); Orange II (86); Metanil Yellow (95); Safranin (584); Methylene Blue (650). (Zts. Nahr. Unters. Hygiene, 1891, v. 5, p. 293.)

### TSCHIRCH.

In 1893 Tschirch recommended as follows:

- 1. The coal-tar colors, and in a narrow sense the anilin colors, are no longer harmful on account of their arsenic content, since at the present time the great majority of them are prepared free from arsenic.
  - 2. Some colors have shown themselves to be harmful to the system.

- 3. Coal-tar colors in general should therefore be permitted for the coloring of foods, but those that have been found to be harmful should be expressly and specifically forbidden.
- 4. The amount of coloring matter which has been determined quantitatively in bonbons and liqueurs is so small that even the ones regarded as poisonous would not be able to develop their harmful effects. (Zts. Nahr. Unters. Hygiene Waarenk., 1893, v. 7, p. 338.)

### KAYSER.

### In 1895 Kayser expressed himself as follows:

As to the poisonous nature of organic coloring, and in respect to their composition, H. Erdmann (*Pharm. Centrall.*, 1892, v. 33, p. 357) concludes that in general acid dyestuffs can pass as nonpoisonous; whereas in the case of basic coloring matters it is recommended to make a physiological examination before using them for the coloring of things in daily use, especially articles of food. Whether that portion of that view which deals with acid dyestuffs will retain unexceptionable and positive validity appears doubtful.

At the present time, speaking generally, interested manufacturers take the point of view that all the coloring matters which are not forbidden in the food law are to be regarded as permitted. Whether this point of view is free from legal objection can not be discussed here; that, however, it can not be accepted from a hygienic point of view under any circumstances whatever does not require any special proof for those conversant with the facts. The hygienic requirements under all circumstances can be summed up in the following rule:

Every coloring matter is to be regarded in every way as suspicious, so far as its harmlessness is not proven by experience or by correct physiological experiments.

No one can say in advance that among the colors which are to-day manufactured and used, which are as yet not called into question, there are none which possess distinctly poisonous properties. Correct examination of artificial coloring matters in this direction is, as is well known, even to the present almost wholly lacking. (Forschungsberichte über Lebensmittel, etc., 1895, Vol. II, p. 181.)

### WEYL.

## In 1896 Weyl expressed himself as follows:

Since the number of the organic coloring matters already known is a very large one, and since their number is increasing daily, and it seems to be unlikely that each individual of these coloring matters will be examined as to its poisonous nature, there are only two ways left in which to solve the question as to the use of coloring matters in the manufacture of food and articles of daily use.

One of them, and at the same time the simplest, would be to prohibit the use of all coloring matters for the coloring of foods, etc. This rigorous point of view will hardly ever be taken by legislators, because it would be tantamount to the removal of many marks of differentiation which have become desirable and necessary.

The second way seems to be the much more practicable, and which Theodor Weyl proposed some time ago.

It consists in permitting the use of only a definite number of coloring matters, recognized as harmless, for the coloring of articles of food, etc. Which coloring matters are to be so permitted is to be determined by the authorities having jurisdiction. The same authorities are also to determine the maximum amount of each coloring matter which can be used for any purpose. New coloring matters can be used only for the above-mentioned purposes when they have been recognized as nonpoisonous after official test. All permitted coloring matters must be also detectable, even in small amounts. (Handbuch der Hygiene, 1896, Vol. III, p. 385.)

### NATIONAL CONFECTIONERS' ASSOCIATION.

In 1899 the National Confectioners' Association of the United States issued an official circular, which has been previously discussed, designating certain colors as harmful, and certain others as harmless; the members of each class are given in the following list, in which only one trade name is given, the Green Table number appearing in parentheses at the end of that name.

### HARMFUL ORGANIC COLORS.

Red colors: Ponceau 3RB (163); Crocein Scarlet 3B (160); Cochineal Red A (106); Crocein Scarlet 7B (169); Crocein Scarlet O extra (164); Safranin (584).

Yellow colors: Picric Acid (1); Martius Yellow (3); Acme Yellow (84); Victoria Yellow (2); Orange II (86); Metanil Yellow (95); Sudan I (11); Orange IV (88).

Green colors: Naphthol Green B (398).

Blue colors: Methylene Blue BBG (650).

Brown colors: Bismarck Brown (197); Vesuvin B (201); Fast Brown G (138); Chrysoidin (17, 18).

### HARMLESS ORGANIC COLORS.

Red colors: Artificial Alizarin and Purpurin (534); Eosin (512); Erythrosin (517); Rose Bengale (520); Phloxin (521); Ponceau 2R (55); Bordeaux B (65); Ponceau 2G (15); Fuchsin S (462); Archil Substitute (28); Orange I (85); Congo Red (240); Azorubin S (103); Fast Red D (107); Fast Red (105); Ponceau 4GB (13); Fuchsin (448).

Yellow and Orange colors: Naphthol Yellow S (4); Brilliant Yellow (5); Fast Yellow (8); Fast Yellow R (9); Azarin S (70); Orange (43).

Green colors: Malachite Green (427); Dinitrosoresorcin (394).

Blue colors: Indigo (689); Gentian Blue (457); Coupiers Blue (600).

Violet colors: Paris Violet (451); Wool Black (166); Azoblue (287); Mauvein (593).

Brown colors: Chrysamin R (269).

### SCHACHERL.

## Schacherl in 1903 made the following statement:

If coal-tar colors are to be permitted for the coloring of food, then, in my opinion, it is not right to limit the use of such to a few coloring matters, but groups of coloring matters must be permitted which are without suspicion from a sanitary standpoint, and which are characterized by definite reactions. Other groups, on the other hand, which contain harmful or merely suspicious colors, must be absolutely excluded. \* \* The selection would be easily made if sufficient data were at hand with respect to the physiological action. Unfortunately this is lacking, a circumstance which need not be surprising in view of the very large number of synthetic coloring matters, since the Schultz-Julius tables enumerate 681 such colors. Unfortunately the experiments of Th. Weyl, which were planned on a large scale, have not been completed, and apart from isolated investigations we are limited in passing judgment upon the most of these colors to the proof that to date nothing with respect to harmful action has become known. \* \* \* Consequently it is still possible that one or the other coloring matter which may to-day be regarded as above suspicion, or a newly discovered coloring matter may possess poisonous properties; the legislators should under all circumstances have the power to exclude for use in foods all suspicious combinations, and all such coloring matters as are not easily distinguishable from them.

The final recommendations of Schacherl amount to permitting—

- 1. All the Azo colors, Nos. 7 to 393 of the Green Tables, except No. 86.
- 2. All the Triphenylmethane colors, Nos. 427 to 492 of the Green Tables, except the hydroxyl derivatives, which would be Green Tables Nos. 483, 484, 485, 486, and 491.
  - 3. All Pyronins, Nos. 493 to 527 of the Green Tables.
  - 4. All Oxyketones, Nos. 537 to 570 of the Green Tables.
  - 5. All Indulins, Nos. 599, 601, and 603 of the Green Tables.
  - 6. Naphthol Yellow S, G. T. No. 4.
  - 7. Methylene Blue, G. T. No. 650.

The use of all other coal-tar colors would best be forbidden, partly from hygienic and partly from practical considerations (rendering control more easy), until the absolute harmlessness of the group in question is determined by physiological test. 
\* \* \* It should be required of all permitted coloring matters that they shall not contain substances which are harmful to health, or even suspicious, either in chemical union or as contaminations. (Fifth International Congress of Applied Chemistry, Berlin, 1903, Vol. IV, pp. 1041–1048.)

The exclusions recommended are all nitro-colors, except Naphthol Yellow S; all acridin colors and all chinolin colors; Auramin, Indophenol and all nitro-colors, except Naphthol Yellow S, Schacherl regards as not necessary; further, he has no knowledge of the physiological action of any of the azoxy or the thiobenzenyl colors, and aside from Methylene Blue, he has no knowledge of the physiological action of the oxazins and thiazins.

### CLASSIFICATION OF RECOMMENDATIONS IN THE LITERATURE.

The following table shows the groups of coal-tar colors of the Green Table classification and the Green Table numbers of the members of each of the groups, together with the favorable or unfavorable recommendations found in the literature in regard to each and a statement as to those regarding which no recommendations are made:

Tabulation of recommendations found in the literature.

G.				er of indi ported or	Total number.		
Group num- ber.	Green Table number.	Name of color.	Un- favor- ably.	Favor- ably.	Conflict- ingly.	Re- ported.	Not reported.
			(1)	(2)	(3)	(4)	(5)
1 2 3	1-6 7-132. 133-336 337-382	Nitro. Monoazo. Disazo. Trisazo.	4 3 3	2 10 3	0 19 7	6 32 13	0 94 191 46
5 6 7 8	383–393 394–398 399–416 417–424	Tetrakisazo. Nitroso. Stilbene Oxyketone.	0 0	1 1	1 0	2 1	11 3 17 8
9 10 11	425–426 427–492 493–527	Diphenylmethane Triphenylmethane Xanthenes	1	0 5 5 0	0 8 1	1 18 8 2	1 48 27
13 14 15	528-533 534-570 571-578 579-616	Anthracene. Indophenols. Azins.	0 2 2	0 1 3	1 0 2	1 3 7	4 36 5 31
16 17 18	617-648 649-657 658-663 664-667	Oxazins. Thiazins. Thiobenzenyl. Ovipalin	2 3 1	0 0 0	0 1 0	2 4 1	30 5 5 3
20	668-688 689-695	Quinolin Sulphids Indigos	2 0	0 1	0 1	2 2	19 5
		Total	33	32	41	106	589

### CONCLUSIONS.

Applying the Schacherl rule, "Other groups which contain harmful or merely suspicious colors must be absolutely excluded," to this table and assuming that all entries in columns 1 and 3 shall be regarded as rendering such colors as "harmful or merely suspicious," it will be found that only one group, namely the Stilbene group, would be permitted under that rule; further, that this rule would admit 17 colors, not one of which has been reported on in the literature as to its physiological action. This state of affairs tends to emphasize the difficulties in the way of any generalization which will be safe so far as public health is concerned and fair to those who use food colors for admittedly legitimate purposes and to make the following recommendation appears to be the only satisfactory way of solving the food-color problem:

Although it would be possible to draw quite reliable conclusions as to the advisability of employing certain colors for food products on the basis of their chemical constituency, the mode of their manufacture and of the ingredients used in same, nevertheless, I think that by far the safest way would be on the one side to force the dealers of colors intended for food products to sell only such colors with which exhaustive and careful physiological tests have been made by experienced and especially impartial and thoroughly reliable people, thereby establishing their harmlessness beyond a doubt. On the other hand, the manufacturers and canners of food products of any description should be forced to purchase and use only those colors which they are sure have been submitted to such careful tests as have been described and by these tests found to be harmless. (Lieber, The use of coal-tar colors in food products, 1904, p. 150.)

This view is confirmed by Santori (Moleschott's Untersuchungen, 1895, Vol. XV, p. 57), who says:

From all these experiments it follows that it is impossible, as some have desired to do, to conclude simply from the chemical composition and constitution whether any given coal-tar dye is poisonous or nonpoisonous. Thus Indulin belongs to the same group as Printing Blue and Methyl Violet to the same group as does Acid Violet; therefore each individual coal-tar dye must be separately examined, and it is only by this laborious method that the use of all really poisonous coal-tar dyes will be prevented.

# VII. RECOMMENDATIONS MADE BY UNITED STATES COLOR INDUSTRIES AND TRADES TO THE DEPARTMENT OF AGRICULTURE.

Prior to the issuance of any regulations, the commission on rules and regulations for the food and drugs act, June 30, 1906, held meetings in New York City during September of that year. The stenographic reports of those meetings, and the briefs filed, in so far as they

<sup>&</sup>lt;sup>1</sup> Santori examined 15 different blue and violet dyes on dogs by the mouth and hypodermically. Of these 15, 8 are poisonous by the mouth and 7 are nonpoisonous by the mouth. He found Indulin to be poisonous and Printing Blue to be nonpoisonous; Acid Violet to be nonpoisonous and Methyl Violet to be poisonous.

relate to colored food, colored food products, or material for coloring foods or food products, have been condensed verbatim in the following pages with the sole exception of omissions, as indicated; for obvious reasons the names of those making the suggestions hereinafter quoted are not given. This review of opinions expressed and recommendations given by the industries and trades most interested in the manufacture, use, and sale of food colors and colored food products is believed to be fair and full with respect to each and every quotation. The numbers in parentheses following each quotation refer to the pages in the stenographic minutes of the hearing from which those quotations are made.

### ANTAGONISTIC TO ALL ADDED ARTIFICIAL COLOR.

- 1. Our position in the coloring question is that we are opposed to all artificial coloring matter in food products. \* \* \* (p. 109).
- 2. Secondly, speaking first of our own business, and I believe that my view would hold as regards all food products, it is my opinion that all added artificial coloring matter in food products should be prohibited. My experience in our line of business demonstrates this to me beyond any question of doubt (p. 439).

### CONCERNING RESTRICTIONS AND REQUIREMENTS.

WHAT CLASSES OF COLORS SHOULD BE PERMITTED TO BE USED.

- 1. All colors, irrespective of their class, whether animal, vegetable, or synthetic, which have been physiologically and chemically examined, and which will neither retard digestion nor have special physiological effects when consumed in quantities corresponding to two grains per day per adult (p. 106).
- 2. On the use of colors we recommend that any kind of a harmless color should be permitted provided it is not a color generally known to be poisonous, or generally found to be poisonous, or one that may be almost impossible to be produced without containing some poison within itself, when finished and ready for use (p. 119).
- 3. Only such colors as are guaranteed to be harmless by reliable manufacturers should be used in the manufacture of confectionery (p. 555).

### WHY COAL-TAR COLORS SHOULD NOT BE BARRED.

- 4. Coal-tar colors, as a class, should not be prohibited; but all those coal-tar colors generally found to be poisonous, or which are hard to produce without containing poisonous properties when ready for use, should be forbidden the privilege of being used or offered for sale for use in food (pp. 116, 117).
- 5. I ought to put in a plea for the use of coal-tar colors, harmless, of course, for the reason that we have not as yet been able to find any vegetable coloring that is suitable that will give us the results that we require. Coal-tar colors, as everybody knows, are much stronger and are more soluble and not acted upon by acids, whereas the vegetable colors, with but one exception, which is a dark red, we have found great difficulty in making use of for bottled soda water. Almost all the vegetable colors either fade out or change on account of the citric acid in the syrup or food, or form in a little while a precipitation which renders the goods unsalable (pp. 119, 120).
- 6. \* \* \* aniline butter colorings \* \* \* are superior to all vegetable colors in the following points: (1) Shade and brilliancy. (2) Strength, by which less foreign material is introduced into the butter. (3) Permanency when exposed to light and cold storage. (4) No effect on the taste or flavor. (5) A clear solution

without sediment or mud which gives the butter a uniform tint without specks (pp. 176, 177).

7. We recommend \* \* \* that the use of harmless coal-tar colors, such as chrysoidine, tropæoline, azoflavine, rocelline, ponceau, Bordeaux, Biebrich red, sulphonated fuchsin and naphthol yellow S be allowed, subject to declaration on the label of the quality and quantity of the color used (p. 226).

### RELATION OF LABEL TO COLORED FOOD PRODUCTS.

- 8. \* \* \* if it seems wise in the opinion of the commission to allow certain coloring matters in food products, then the names of the coloring matter ought to be stated on the article (p. 109).
- 9. \* \* I suggest that the use of aniline colors should be made proper if it is so stated on the label (p. 127).
- 10. We recommend \* \* \* that the use of harmless coal-tar colors \* \* \* be allowed subject to declaration on the label of the quality and quantity of the color used (p. 226).

### LABELING OF FOOD COLORS AS DISTINGUISHED FROM COLORS FOR OTHER USES.

11. We recommend that if by any means the Government has the power under the food laws to compel color manufacturers so to do, they be compelled to label all packages containing colors intended to be used in articles of food as colors intended for such purposes as distinguished from colors intended for other purposes; \* \* \* (pp. 555, 555a).

### THE TEST OF A HARMLESS COLOR.

12. A harmless color is one "which will neither retard digestion nor have special physiological effects when consumed in quantities corresponding to two grains per day per adult" (p. 106).

### QUANTITY OF COLOR TO BE CONSIDERED IN DETERMINING HARMLESSNESS.

- 13. The quantity of 2 grains is mentioned here because in confectionery where these harmless colors are more used perhaps than in any other product, it would be a proportion of one part of color to 3,500 parts, representing 1 pound of color to 3,500 pounds of confectionery, and that is why that was accepted, because that is practically the maximum quantity used in confectionery (p. 106).
- 14. In the very minute quantities in which the colors are used in carbonated beverages, it would seem a great hardship to prevent us from using coal-tar colors. One ounce of coal-tar red will color satisfactorily from 1,000 to 2,000 gallons of soda water. Of yellow and orange 1 ounce will color from 1,000 to 8,000 gallons. It is readily seen that unless the coloring used is absolutely a violent poison it can have absolutely no effect on the consumer who takes it in an 8-ounce glass, and who could not possibly consume half a gallon or a gallon of that product (p. 119).
- 15. On this basis 1,000 pounds of butter would contain 420 grains of aniline color (p. 183).
  - 16. One ounce of color to 30 pounds colored food (p. 135).

### THE NUMBER OF DIFFERENT COAL-TAR COLORS REQUIRED.

17. And if I were on this committee I would advocate taking out two or three dyestuffs which beyond any shadow of doubt are harmless, and which have been experimented upon, and which would be sufficient for all the purposes of the industry \* \* \* none of these colors are patented, anybody can manufacture them (pp. 147, 148).

18. You have been told that the food commission of the State of Pennsylvania is going to rule that seven colors shall be allowed in the State of Pennsylvania. That is not enough, because it is not possible to reproduce all of the required shades with those colors unless you take the seven primary colors, when you can reproduce any colors. But it is absolutely necessary to have about three yellows to meet the requirements of the trade. Some of the articles that are put out have an organic acid nature, and the anilines will stand that. \* \* \* On that account the character of the food product must be taken into account in the use of the color, and the confectioners have about 30 colors that I know positively of \* \* \* (p. 160).

19. We recommend that \* \* \* chrysoidine, tropaeoline, azoflavine, rocelline, ponceau, Bordeaux, Biebrich red, sulphonated fuchsin, naphthol yellow S be allowed \* \* \* (p. 226).

### MANUFACTURING REQUIREMENTS WHICH COAL-TAR COLORS MUST FULFIL.

- 20. Coal-tar colors, as everybody knows, are much stronger and are more soluble and are not acted on by acids. \* \* \* Almost all the vegetable colors either fade out or change on account of the citric acid in the syrup or food, or form in a little while a precipitation which renders the goods unsalable (p. 119).
- 21. There are many of these colors (coal-tar colors) that will not stand the natural acids produced in manufacturing foods. For instance, in making confectionery, in boiling candy you make a certain grade of candy where the mixture is boiled to 230°. A certain color will stand that temperature. Then you take another candy and that is boiled to 320° or 340° F., and the colors that will stand 230° will in many cases not stand the temperature of 340°, while another class of colors will stand that temperature. So you have got to distinguish and get a color that will stand these difficult requirements (pp. 159, 160).

### GUARANTEES AND GOVERNMENT CONTROL.

- 22. Only such colors as are guaranteed to be harmless by reliable manufacturers should be used in the manufacture of confectionery (p. 555).
- 23. We recommend \* \* \* that the Government procure samples of such colors from time to time, wherever they have jurisdiction so to do, and if such colors be not legal for such purposes under the food law, that the same be prosecuted and driven off the market in so far as the Government has the power to do so; and if no means can be devised to compel such labeling of colors intended for food purposes, then that the Government procure such evidence as possible as to the purpose for which a color is intended to be used, and if such evidence shows a food purpose, that such color be prosecuted if illegal under the food law (pp. 555, 555 a).
  - 24. See page 13.
- 25. The chemical test is the first, and that might throw out a color on account of its containing a little tin or zinc, or some substance foreign to the food product. Whether that is deleterious in the quantity in which it is present or not is immaterial (p. 160).
  - 26. In a brief filed the following suggestions were made:

It must be stated that all the chemist can determine is whether or not the colors contain some impurities that are known to be of poisonous nature. Aniline colors are or can be made entirely free of such impurities, and with this fact established the task with the chemist is exhausted. When it comes to decide the question whether or not a color by itself, when free of all impurities, is injurious to health or not, then the chemist is not the proper authority; it is for the physiologist and for the medical profession to pass on such questions. Chemical theories go for nothing in deciding such questions. It would not even do to classify colors or other substances according to their makeup, as it has been shown again and again that substances belonging to the same chemical class are entirely different in regard to the physiological conduct.

Reliable information on this subject can be gained only by physiological experiment, as we can not say definitely whether a substance, color or any other, is injurious or not without finding out for every substance by experiment. This has been done for a considerable number of aniline colors, and these experiments are the only things that deserve any attention. Everything else is idle talk. \* \* \* There are a very large number of aniline colors that have not been treated yet, and we are safe in saying that among these will be also some harmless and others injurious. As they have not been experimentally tested, we do not know which are harmless and which are not; It will therefore be clear that a law forbidding the indiscriminate use of aniline color for the purpose of coloring articles of food is necessary and useful.

But if the meaning of the law is to prevent only the use of injurious colors, as it appears to be, then the way to proceed would be very definitely outlined. Besides physiological colors, all such colors should be forbidden that have been found to be injurious and such aniline colors as have not been tested sufficiently. There will then be left over a number of aniline colors which have been proven by experiment

to be entirely harmless, even if taken in large doses.

The experimentors were quite impartial. They had no preconceived ideas, but started simply to find out the true state of affairs. Their reports are therefore very reliable, and it will not do to overlook or to ignore them. The colors that they found harmless can be considered perfectly safe, so much the more as the doses conveniently taken with food would be much smaller than the doses that have proven to be harmless. These harmless aniline colors carry all the shades wanted in the food industry. The law should provide that one of these colors (or mixtures of these) must be used when a food article is being colored, because these few aniline colors are the only colors that can be considered perfectly safe as far as our present knowledge goes. Nothing should be left to guesswork or experimenting, as is the case just now. The colors that are permitted should be enumerated by their scientific as well as by their commercial names, and only such colors should be listed as permissible for coloring food products as have been proven to be harmless, even in large doses. Provision should be made to insure the purity of the colors sold for coloring food; the manner of packing such colors and the labeling of same should be laid down clearly, and all colors now listed, aniline as well as physiological colors, should be strictly forbidden. If the problem is viewed without preoccupation and prejudice, the facts given above will speak for themselves.

# VIII. INVESTIGATIONS, OTHER THAN ON ANIMALS, BEARING ON THE HARMFULNESS OF COAL-TAR COLORS.

### PFEFFER.

Pfeffer, writing on the Absorption of Anilin Colors by Living Cells, summarizes his results as follows:

The relatively little poisonous Methylene Blue does damage protoplasm in a solution of  $0.001~\mathrm{per}$  cent.

Methyl Violet: This coloring matter is not only stored up in the juices of the cell, but is also able to color the living protoplasm, and care is necessary, on account of the poisonous nature of the Methyl Violet, to prevent damage; these cautions are based upon solutions of 0.0003 to 0.00001 per cent strength.

Methyl Violet less poisonous than Cyanin.

Bismarck Brown about as poisonous as Methylene Blue.

Fuchsin as poisonous as Methylene Blue.

Safranin as poisonous as Methylene Blue.

Methyl Orange is poisonous only to a slight degree.

Tropæolin 000, Tropæolin 00, and Rosolic Acid are not poisonous.

Methylene Green as poisonous as Methyl Violet.

Nigrosin as poisonous as Methyl Violet.

Eosin (Tetraiodo fluorescein) kills in 0.1 per cent solution, but lets live 24 hours in 0.01 per cent solution. (*Untersuchungen aus dem Botanischen Institut zu Tuebingen*, 1886-88, vol. 2, pp. 179-331.)

### WINOGRADOW.

Winogradow reports on the influence of certain coal-tar colors on digestion, which experiments were carried out in glass. The conclusions arrived at are as follows:

The twelve colors, Safranin (584), Azo Fuchsin G. (93), Coerulein S. (527), Jodeosin (516), Magdala Red (614), Benzopurpurin (277, 278, 279, 307), Ponceau 2R (55), Orange II (86), Phloxin RBN (?), Chrysanilin (532), Azoflavin (92), and Cerise (mixture of 448 and 532), even in amounts of a few milligrams, which in relation to the digestive fluid make up only a few tenths or hundredths of a per cent, exercise a strongly retarding, almost completely inhibitive, action upon the peptic digestion of albumen.

The thirteen colors, Chinolin Yellow (667), Acid Green (434, 435), Azo Acid Yellow (92), Naphthol Yellow (4), Primulin (659), Anilin Orange (87), Metanil Yellow (95), Methylene Green (651), Iodin Green (459), Yellow T (84), Anilin Green (?) Auramin O (425), and Martius Yellow (3), retard the digestive action noticeably, although to a slighter degree than the first 12 colors; in any event they are not indifferent. (Zts. Nahr. Genussm., 1903, v. 6, p. 589.)

### HEIDENHAIN.

Heidenhain, in his book entitled "Ueber chemische Umsetzungen zwischen Eiweisskorpern und Anilinfarben" (Bonn, Germany, 1902), reports on the behavior of 70 different coal-tar colors, 3 intermediate products, and 4 raw materials toward various albuminoids such as serumalbumen, albumen, and casein.

Of these 70 colors, 21 have been investigated physiologically, and the results embodied in this report; and of these 21, 17 were on the United States market as food colors and 4 were not on this market.

Limiting the attention to the 17 that were offered, 4 of them are among the 7 permitted colors of Food Inspection Decision 76.

In the cases where albumen and casein were used, they were employed in 0.5, 0.1, 0.02, 0.01, and 0.005 per cent solutions, acidified with acetic acid; the coloring matter was employed in a 1 per cent solution in the first strength, in 0.1 per cent solution in the second and third strengths, and in a 0.02 per cent solution in the fourth and fifth strengths, and one volume of coloring-matter solution was brought in contact with five volumes of albuminoid solution.

The following colors precipitated the albuminoid in all the strengths. The numbers in parentheses indicate the Green Table numbers;

<sup>&</sup>lt;sup>1</sup> It is not always possible to identify the trade names given by Winogradow with specific numbers in the Green Tables; the number in parentheses after the name indicate the number in the Green Tables wherever that identification could be made with any reasonable certainty; wherever two or more numbers appear, the context indicates that the name might apply to any one or all of them.

the numbers in italics are those of the permitted list of Food Inspection Decision 76: (55) Ponceau 2 R, (65) Fast Red B, (434) Light Green SF bluish, (56) Ponceau 3 R, and (106) New Coccin.

The following precipitated in all but the fifth strength: (107)

Amaranth.

The following precipitated only the first three strengths: (14) Orange G and (85) Orange I.

The following precipitated only the first two strengths: (462) Acid

Magenta.

For the basic colors the method of testing was different from that described for the acid colors and the amounts employed were not so definitely set forth. A 1 per cent solution of serum albuminoid was employed; the solution of coloring matter used varied in strength from 0.5 to 1 per cent (b), a "very dilute solution" (a) being also employed. The annotations given by the author (p. 114) are herewith reproduced in full:

(17) Chrysoidin Y. (a) In a very dilute solution the yellow base is at once liberated, and when sufficient color is added albumen precipitation takes place. No color change on heating. (b) Turns yellow at once. Further additional color produces a brown and albumen precipitation.

(201) Manchester Brown. (a) Becomes yellow at once, due to separation of the free base. On heating no change. (b) Becomes a discolored brown, and produces a

nice brown albumen precipitate.

(425) Auramin O. (a) No change. (b) Precipitates albumen strongly.

(427) Malachite Green. (The oxalate of the color was used.) (a) Color changes from a blue-green to a more pure green. (b) Cold, no precipitation of albumen; heated, sudden precipitation of albumen.

(428) Brilliant Green. (A sulphate of the color was used.) (a) Becomes milky.

(b) Immediate precipitation of albumen.

- (448) Magenta. (Acetate and nitrate were used, and in both acted the same.) (a) Color changes from yellowish-red to rose-red. (b) Albumen precipitation abundant, even in the cold.
- (451) Crystal Violet. (a) The color loses its reddish cast and changes to pure blue.

(b) Albumen precipitated by the use of much color.

(4) Naphthol Yellow S was tested as the free-color acid and not as the sodium or potassium salt, which is its commercial form. It precipitated the albuminoid from the following solutions: 1. 1 per cent serum albumen in water. 2. 0.5 per cent serum albumen in 10 per cent acetic acid. 3. Casein, 0.5 per cent in 10 per cent acetic acid. 4, Serum albumen; 5, Casein; 6, Nuclein, all in 0.5 per cent solution in 0.2 per cent sodium hydroxid. 7. Nucleinic acid in 0.5 per cent water solution.

The relationship between the amounts of color-acid and albuminoid solution here used does not appear definitely in the book.

Having reference now to the literature and the physiological action, as compiled herein, it will be observed that the ability to precipitate albumen, or not to precipitate it, under the conditions of Heidenhain, appears not to have any direct connection with the results obtained by actual physiological test on animals or man. For instance, among the five that precipitated all five strengths of

albumen and casein, No. 65, Fast Red B, of the Green Tables, has been found to be not harmful by tests actually described; No. 56, Ponceau 3 R, belongs to a class of colors generically permitted by the law of Austria; Nos. 55, Ponceau 2 R, and 106, New Coccine, have been reported on both favorably and unfavorably by different experimenters; No. 434, Light Green SF bluish on physiological examination has been described as suspicious.

From this it appears that two colors, physiologically probably harmless, precipitated all the five strengths of albumen, and three colors, which are perhaps no more than suspicious, likewise precipitated all five strengths of albumen.

No. 107, Amaranth, which has been examined with favorable results by two different experimenters, precipitates four out of the five strengths.

Of the two colors precipitating the first three strengths, both have been examined physiologically with favorable results, namely, Nos. 14, Orange G, and 85, Orange I.

The color which precipitated only the first two strengths, namely, No. 462, Acid Magenta, has likewise been examined physiologically, and the reports are favorable.

Thus it would seem that there is no definite connection between the physiological action and the ability to precipitate albumen from acetic acid solution in the case of acid colors.

In the case of basic colors the situation seems to be somewhat different.

### OTHER AUTHORS.

Rosenstiehl (Fifth International Congress of Applied Chemistry, Vol. III, p. 700) states that when the color is present in an excess, yeast absorbs 8 per cent of Magenta (448) and 5 per cent of Malachite Green (427, 428). The Acridins (528-538), the Thionins (649-657), the Safranins (583, 584), and the Rosanilins (447-448) dye yeast the best; solutions containing 3 per cent by weight of the dry weight of the yeast are completely decolorized by such yeast and at ordinary temperatures. The Eosins and Phthaleins dye the yeast only incompletely, whereas Azo dyes (7-393) (with the exception of Benzo Purpurin, 277, 278, 279, 309) do not dye yeast at all. Such dyed yeast, however, is not dead; it has merely lost its power to cause fermentation. The numbers in parentheses are the corresponding numbers in the Green Tables as nearly as they could be identified.

Bokorny (*Chem. Ztg.*, 1906, v. 30, p. 217) examined Magenta (448), Safranin (584), Victoria Blue (487, 488 or 490), Methylene Blue (650), and Alizarin Blue (562 or 563) (the numbers in parentheses are the probable Green Table numbers) as to their effect on micro-organisms such as yeast cells, infusoria, and the like, and found that these dyes

in a concentration of 1:100,000 killed them, whereas strychnin nitrate in the same concentration is substantially without effect. Death is caused by absorption of the dye by the albumen of the protoplasm. The dyes seem to be absorbed not only by living albumen, but also by living nerve cells and fibers. Pure anilin or coaltar colors, however, are not poisonous in the ordinary meaning of the words, that is, humans are not likely to be easily injured by them.

Houghton (J. Amer. Chem. Soc., 1907, v. 29, pp. 1351-1357) shows that Bismarck Brown (197, 201) and Crocein Scarlet (160?) hinder

the peptic digestion of fibrin, casein, and albumen.

Stilling (Anilinfarbstoffe als Antiseptica, 1890, v. 2, pp. 55-56) states that he found the animal cells to be affected by pure coal-tar colors in the same way that vegetable cells were affected.

Penzoldt, based upon the experimental work of Beckh (Archiv. Exp. Path. Pharmak. 1890, v. 26, p. 310), reports as follows:

Of the 15 dyes-

9 TO	dyes	
	Name.	Green Table number.
1.	Malachite Green	427
2.	Hofman's Violet (methyl variety)	450
	Methyl Violet.	
4.	Rose Bengal (Erythrosin)	517
	Phenyl Blue	
6.	Methylene Blue.	650
	Fuchsin	
8.	Coralline	484
9.	Eosin	512
	Methyl Orange.	
11.	Vesuvin	197
12.	Tropaeolin	88
13.	Scarlet Red	(?)
14.	Congo Red	240
15.	Indulin sulpho acid	601
	•	

all of which are water soluble and when used were free from arsenic, only the first six in saturated solution arrested the development of *Staphylococus pyogenes aureus*, and of these six all but the Erythrosin and Methylene Blue arrested the growth of anthrax bacillus.

Of these six when injected into rabbits subcutaneously the following produced no changes of consequence: Erythrosin (250), Phenyl Blue (100), Methylene Blue (75). The numbers in parentheses show the number of milligrams of dye per kilo body weight of the rabbits.

Methyl Violet (50) produced only local changes, such as gangrene.

Malachite Green (100) and Hofman's Violet (20) produced muscular paralysis, which in the case of Malachite Green resulted fatally on the ninth day; in the case of Hofman's Violet the paralysis was complete on the tenth day.

The remaining nine colors are apparently of no effect upon staphylococus or upon anthrax.

H. W. Williams (A Manual of Bacteriology, 1906, p. 200), under "Disinfectants and Germicides," says:

Aniline dyes. Many of these substances, notably pyoktanin (Methyl Violet), possess germicidal properties. A solution of 1:5000 will kill the anthrax bacillus in two

hours. A much stronger solution, 1:150, is required to kill the typhoid bacillus in the same time. Malachite Green is said to possess even greater germicidal power than pyoktanin. Methylene Blue also possesses considerable germicidal power.

# IX. COMPILATION UNDER THE GREEN TABLE NUMBERS OF ALL INFORMATION AVAILABLE AS TO THE SUITABILITY OF COALTAR COLORS FOR FOOD.

### GENERAL STATEMENTS.

Before entering upon a detailed study of what has been published for and against specific coal-tar colors, it is probably well to consider, for whatever they may be worth, some of the general statements that have been made, from time to time, in the literature relative to coal-tar colors, considered either as a whole or as subdivisions or classes thereof, and their physiological action or their suitability for use in foods.

1. Schultz (Die Chemie des Steinkohlentheers, Brunswick, 1887–1890, Vol. II, p. 35), after discussing the regulations of the German Empire in respect to food coloring, says:

With respect to these regulations the artificial organic coloring matters can be regarded, in general, as harmless. For the purposes of dyeing magenta made by means of arsenic acid, further picric acid, and those coloring matters which occur as oxalates or zinc chlorid double salts, such as Methylene Blue and Bitter Almond Oil Green, can be used. The use of the substances named for the coloring of food products is, however, suspicious and should not be permitted.

2. Stilling (Anilinfarbstoffe als Antiseptica, Strassburg, 1890, Vol. II, pp. 55-56) says:

In view of the fact that the most innocent substance, such as distilled water or common salt, when introduced in large quantities into the organism can act fatally, the anilin coloring matters therefore, particularly if they be free from all admixtures, such as arsenic, copper, and chlorid of zinc, are to be regarded as wholly nonpoisonous.

All experience gathered since my first publication has likewise fully confirmed this nonpoisonous nature.

3. Stilling (Ber. Klin. Wochensch. 1890, p. 531) also says:

Proceeding from purely theoretical views, and based upon these botanical and physiological experiments, I have recommended anilin colors as antiseptics for the following reasons:

1. They are to be designated as absolutely nonpoisonous. This will be confirmed by every chemist acquainted with the relevant details, and also from the medical side this has long ago been determined by Grandhomme. The publications of this author, who made extensive observations and experiments in the anilin factory of Meister, Lucius & Bruening, appeared in the beginning of the eighties, and has hardly become known in medical circles. However, I was first made acquainted with this by my colleague, Prof. Rose, in Strasburg.

2. That it is possible to bring about death in experimental animals by introducing large amounts of the coloring matter into the peritoneal cavity proves nothing against

the nonpoisonous nature of these substances. This action is to be regarded as a purely mechanical one, a view which I will thoroughly confirm in my second communication.

With respect to the anilin colors not soluble in water, Ehrlich long before me, in his excellent publication on the oxygen requirements of the organism, has arrived at this view, and has excellently described it as merely a penetration of the organs.

# 4. Lehmann (Methoden der Praktischen Hygiene, Wiesbaden, 1890, p. 543) says:

The hygienic significance of coal-tar colors has heretofore been judged quite variously. When the intensely poisonous nature of the first impure and particularly arsenic-containing coloring matters became known the inclination was to judge the coal-tar coloring matters very strictly; when it was subsequently recognized that the contaminations were largely the cause of the harmfulness to health, there followed a period in which no poisonous coal-tar coloring matter was known in any pure condition. (Eulenberg & Vohl, 1870.) More recent investigations, however, have disclosed a series of coal-tar coloring matters which, as a matter of fact, possess a considerable poisonous action, and already cases, although not numerous, have become known in which serious and even fatal poisonings by means of pure coal-tar colors have arisen. Alongside of this there still continue to exist the possibilities described by Eulenberg and Vohl (Viertel-Jahressch. für Gerichtliche Mediz, 1870), whereby harmless coloring matters become harmful; but the realization of these possibilities has become essentially more veldom through improvements and changes in manufacture.

5. Stilling (Arch. Exper. Pathol. Pharmak., 1891, v. 28, p. 352), in speaking of the anilin colors as antiseptics, says:

It is the nonpoisonous nature of these substances, their easy solubility and diffusibility, and above all their inability to coagulate albumen which lends them their importance, which now can be only difficultly denied.

Note.—The work of Heidenhain abstracted in Section VIII does not fully bear out this article.

6. Erdmann (*Pharm. Centrall.*, 1892, v. 33, p. 357) says:

The sulphonated as well as the carboxylated coal-tar dyes will not have any pronounced action on the organism. Acid dyes may therefore be regarded in general as nonpoisonous, whereas in the case of basic dyes a physiological examination is to be recommended before they are permitted to be applied to articles in daily use or indeed to be used in food or drink.

Note.—Out of the 80 different dyes on the food-color market in the summer of 1907 whose composition was avowed, 15 were basic and 65 were acid.

- 7. Tschirch expressed himself as follows:
- 1. The coal-tar colors, and in a narrower sense the anilin colors, are no longer harmful on account of their arsenic content, since at the present time the great majority of them are prepared free from arsenic.
  - 2. Some colors have shown themselves to be harmful to the system.
- 3. Coal-tar colors, in general, should therefore be permitted for the coloring of foods, but those that have been found to be harmful should be expressly and specifically forbidden.

- 4. The amount of coloring matter which has been determined quantitatively in bonbons and liqueurs is so small that even the ones regarded as poisonous would not be able to develop their harmful effects. (Zts. Nahrs. Unters. Hygiene Waarenk., 1893, v. 7, p. 338.)
- 8. Georgievics (Lehrbuch der Farbenchemie, Leipzig, 1895 p. 10) says under "Poisonous nature of the coloring matters:"

It is a little known fact that of the very large number of organic coloring matters only a few have been found to be poisonous; these are Picric Acid, Victoria Orange (Saffron-surrogate), Aurantia, Metanil Yellow, Orange II, and Safranin. The prejudice which is still quite widely accepted that most of the artificial coloring matters are poisonous dates from the early periods of anilin-color manufacture, at which time magenta and the coloring matters made from it occurred in commerce highly contaminated with arsenic. At present, however, these coloring matters are prepared absolutely free from arsenic, and are, as such, nonpoisonous. A few coloring matters which occur commercially as zinc chlorid double salts, such as Methylene Blue and Malachite Green, may be harmful in consequence of their zinc content, and should therefore never be employed in the coloring of food products. \* \* \* In consequence of their physiological activity some coloring matters are employed as medicines, indeed principally Methyl Violet, Auramin and Methylene Blue. The first two, known as blue and yellow Pyoctanin (pus-destroying) are, owing to their great antiseptic action and diffusibility, valuable medicines; on account of the unpleasant coloring effect accompanying them they are but little used. Methyl Violet was first recommended as an antibacterial remedy in diseases of the eye by Stilling; subsequently it has been employed in other special cases; its principal use, however, is in surgery for the prevention of malignant proud flesh.

The use of Auramin is entirely analogous.

Methylene Blue (as a free base) is used principally as an analgesic (pain-relieving remedy) and is given internally; on account of its ability rapidly to diffuse through the tissues of the nervous system; it can also be introduced by injection. It is used as a remedy against malaria, carcinoma, Bright's disease, etc.

The following have been tested as remedies or as antiseptics: Safranin, Lydin (Mauvein), Vesuvin, Anilin Blue, Carbolic Magenta, Alizarin Yellow C (Gallacetophenone), etc. The potassium salt of Dinitro-ortho-cresol was brought into commerce by the Farbenfabriken vorm. Friedr. Bayer & Co., under the name of Antinonnin, and has given excellent results as a means against formation of mold in cellars and against wood fungi.

## 9. Weyl (Handbuch der Hygiene, 1896, p. 378) says:

A few organic coloring matters, but only a very few, possess poisonous properties. A rule by means of which the poisonous or nonpoisonous nature of organic coloring materials can be determined without experiment is unknown even for those coloring matters whose constitution has been determined and experiments on the poisonous nature of organic coloring matters are very few in number.

## 10. Lewin (Lehrbuch der Toxicologie, 1897, p. 230) says:

In the use of various fabrics or of foodstuffs, which are colored with anilin or coal-tar colors, or in commercial contact with such colors, local and general symptoms of poisoning, such as eczema, swelling of the face, vomiting, diarrhea, anæsthesia, paresis, etc., have been observed. These are generally due to the toxic nature of the coloring matters, frequently to harmful ingredients of the same, and hardly ever to poisonous mordants. Many workmen in anilin factories show permanent spots; for example, on the cornea and conjunctiva, head, chest, face, and neck, without

any interference with their general condition. Local changes of more serious nature have more frequently been observed in the mucous membranes and on the skin. Thus, in one case, a camel's-hair pencil, soaked with anilin color, accidently entered the eye, and at first nothing was noticed but a violet blue coloration, later inflammation and chemosis took place. I have observed local swelling and indurations of the skin, particularly on the cheeks, in the case of children after they had worn caps colored with anilin dye.

11. Winton (Connecticut Agric. Exper. Sta. Report, 1901, p. 181) says:

Although there is evidence that most of the coal-tar dyes are not injurious to some of the lower animals, it is not safe to assume that they are entirely harmless to human beings. The dog, an animal used in most of Weyl's experiments, has a proverbially strong stomach, and eats with no apparent discomfort many things which would disturb the digestion of a man.

12. Chlopin, in his book published in 1903 (see p. 75), states as follows:

(Page 114.) \* \* \* All the dyes examined by me I divided into three categories: Dyes which caused striking general symptoms of poisoning and led to the death of the animal, or would have led to it if the experiments were not purposely discontinued, I designate by the term poisonous; dyes which induced some separate and temporary symptoms of disease, for instance, vomiting, diarrhea, separation of albumen in the urine, the general condition remaining normal, I designate as suspicious; lastly, the dyes which caused no apparent disturbance during the experiments are designated by the term nonpoisonous. I purposely do not call the last category harmless, because by our experiments the question could not be decided negatively as to whether the nonpoisonous dyes did not cause some finer pathological changes in the organism and functions which could not be detected by simple observation.

(Pages 219-221.) Thus, according to all the investigators quoted, there were found altogether 22 poisonous and harmful dyes, out of about 60 dyes examined; which makes 36.7 per cent of poisonous and harmful dyes among those examined.

My investigations gave 30 per cent of poisonous and 40 per cent of suspicious dyes. The percentages above given have a fairly well established basis, since they were obtained by the examination of 100 dyes, which is about one-fifth of all the dyes in commerce. Further, considering the distribution of the poisonous and harmful dyes according to various chemical groups, we find that they occur in 12 of the 18 groups, and we can not note any regularity in this distribution; it is impossible to say that there is any definite connection between the fact that the dye belongs to a certain chemical group and its action on the animal organism. Usually among the dyes of one and the same group there are some harmful ones, but there are also some harmless ones, and the ones and the others have very similar composition. This or that action of the dyes on the animal organism, as we shall presently see, is determined more by the delicate difference in the internal structure of their molecules than by those differences on which is based, at the present time, the classification of the aromatic dyes.

On the basis of the whole experimental material on hand—mine and that of other investigators—I can make only the following very few and purely empirical generalizations:

1. According to the shade produced, the poisonous and harmful colors are distributed as follows: Most of all poisonous dyes are found among the Yellows and the Oranges; then come the Blues, then the Browns and the Blacks; there are very few harmful dyes among the Violets and Greens; among the Reds was found only one suspicious one, and no poisonous ones.

2. The most poisonous dyes belong to the Nitro, Azo, Triphenyl, and Thiazin groups, and also to the Auramins.

3. A whole group of poisonous and harmful dyes is formed by the new sulphid dyes

known as Vidal's dyes.

(Page 224.) By the facts and observations quoted above is corroborated the opinion that in general the coal-tar dyes, according to the composition and properties, appear as substances foreign to the animal organism, and may influence harmfully the vital functions, even in those cases when they do not possess distinctly poisonous properties. For this reason many hygienists make it a principle not to allow the coloring of food products or of beverages with coal-tar dyes, independently of the fact whether they prove in actual experiments on animals poisonous or not.

We must therefore agree with M. Rudner that food of the masses require the most

far-reaching protection, maintaining them free from foreign additions.

13. Koenig (Die Menschlichen Nahrungs- und Genussmittel, Berlin, 1904, Vol. II, p. 462) says:

Even though the majority of the anilin coloring matters, in view of the small amounts in which they are generally employed, can not be regarded as directly harmful to health, yet the objections to their use in the coloring of food products for the purpose of substituting or strengthening a natural color lies in the deception connected therewith \* \* \*.

### 14. Fraenkel (Arzneimittel-Synthese, Berlin, 1906, p. 570) says:

It is clear that the coloring property of these chemical substances stands in no relation to their physiological actions, but, on the contrary, the physiological actions depend only upon the general structure of these substances, and therefore upon their membership in definite chemical groups.

(Pages 574-5.) We see, even in considering this group of substances, that they do not possess any specific action, but they are capable of use, preferably by external application, as antiseptic materials, as materials which in their action stand somewhere between carbolic acid and corrosive sublimate, and whose coloring property, in consequence of which they were primarily selected, is directly a hindrance in this use, since the coloring of the bandages and the hands of the operators and the skin of the patients certainly can not be regarded as a pleasant occurrence; that the antiseptic action stands in some relationship to the properties of the substances as coloring matter must be positively contradicted. It depends only on the general structure of the substance, and does not stand in any direct relation to the chromophore and auxochrome groups of the substances, but more closely to the aromatic nucleus. Indeed, it may happen that an auxochrome group diminishes the antiseptic activity of such a substance.

Note.—The dyes referred to belong to the Monazo, Disazo, Triphenyl-methane, Xanthin, Azin, and Thiazin classes.

(Page 92.) The investigations of Ehrlich have shown that basic dyes color the brain gray, and, moreover, they color nerve fiber very well, and are therefore to be regarded as neurotropes. The dye acids, on the other hand, do not dye nerve fiber, and in particular the substituted sulphonic acids do not dye tissue at all.

15. Meyer, in his paper on "A preliminary communication on the toxicity of some aniline dyestuffs" (J. Amer. Chem. Soc., 1907, v. 29, p. 892), says:

(Page 893.) "A manufacturing confectioner of this city, for whom I made examination of colors used by him, informs me that a yellow color sold as Auramin has such

high tinctorial power that 1 ounce will color 2,000 pounds of candy to the highest yellow tint required in his business. It is obvious that the toxicity of such a body would have to be very high to render it harmful in such use." Conclusions of this kind do not take into account the possible detrimental action ensuing on healthy as well as diseased persons from long-continued use of small quantities of foreign substances.

(Page 909.) The same author raises objection to feeding experiments on the ground that substances are thereby introduced greatly in excess of the amounts generally found in foods and that the ill effects "are liable to be due to the excess and in long-continued experiments due to a cumulative action of the excess." Surely if excessive amounts have a cumulative action, small amounts may also finally show toxic effects due to retention and accumulation of the poison. To declare a substance entirely innocuous would require evidence as to its nontoxicity both to normal and diseased persons after its long-continued administration in both small and large doses. The most extreme contingencies would have to be provided for. The above objections to feeding experiments are therefore not valid. It is hoped that a study of the effects on metabolism of some of these substances will help to further elucidate the subject.

He summarizes the results of his physiological investigations of seven different coal-tar colors as follows:

- 1. Several commercial organic dyestuffs (Curcumin S, Tartrazin, Naphthol Red S, Carmois in B, Naphthol Yellow S, Gold Orange, and Ponceau 2 R) were studied as to their general effects on dogs when administered in varying amounts and during fairly long periods (two weeks).
- 2. None of these dyestuffs under the conditions above indicated exhibited any marked degree of toxicity. There was only one fatal result, which may have been due to influence independent of the action of the colorant.

Similar quotations from the literature could be added to the foregoing, but these are beyond question sufficient to show that a wide divergence of opinion as to the harmless or harmful nature of the coal-tar colors as a class does exist among scientific men, and that all those above quoted agree that there are some at least of the coal-tar colors which even in a pure state may be harmful to human health, and that the question of actual harmfulness under the conditions of actual use in foods and the consumption of foods is regarded by some as being properly answered in the negative and by others as being properly answered in the affirmative. The question of amount of color employed in the food products and the amounts of such food normally eaten are therefore raised by some as the deciding factors.

In this connection the following statement from page 49 of the arguments before the Committee on Patents in the House of Representatives, April 8, 15, 16, 22, 29, 1908, may be of interest:

\* \* It should be remembered that after a new chemical has been discovered and patented it requires as many as three years of experiment before we dare offer it in this country as a medicine for human beings. These experiments are conducted abroad before we receive it here. It is first tried on animals and gradually, with great caution, extended to human beings in the foreign hospitals, so as to ascertain its physiological effects quantitatively upon the various organs, both when those organs are in the healthy state and when they are affected by various disorders \* \* \*.

A search of the literature herein compiled fails to disclose any such searching physiological examination of any of the coal-tar colors recommended for use by human beings in food products, as is asserted in the above quotation to be necessary in the case of a new chemical intended for use as a drug. If Fraenkel, as quoted on page 60, is correct in his statement that coal-tar colors act physiologically because they are chemicals and not because they are coloring matters, then coal-tar coloring matters prior to use in foods, in which they are used by the young and the old, the well and the sick, without restriction and without supervision, should also be thoroughly tested, and very few, if any, coal-tar colors seem to have been examined with the thoroughness set forth in the above quotation. That uniformity and purity of product is necessary in order to be sure that the chemical is going to act physiologically in the same way every time is obvious. According to Fraenkel, what is true of a chemical is just as true of a coal-tar color, and if uniformity of strength and cleanliness of product are desirable when a chemical is to be used as a medicine, such properties are equally desirable when a chemical is to be used as an ingredient in food.

### CLASSIFICATION OF OPINIONS IN LITERATURE AND IN LEGAL ENACTMENTS SHOWING CONDITION OF THE MARKET IN 1907.

The literature and legal enactments hereinafter grouped under the relevant Green Table numbers have been classified as (1) unfavorable—i. e., only unfavorable reports found in the literature; (2) favorable, and (3) contradictory reports, as each case required; so that under each Green Table entry there is not only the relevant literature, but also the character assigned to it for the purpose of coming to a conclusion as to the propriety of the use of such color in foods as based on such literature, which conclusion formed in that respect the basis for Food Inspection Decisions Nos. 76, 77, and 106.

However, it does not follow that all dyes placed in the "favorable" list are actually harmless; the investigations or opinions reported of each may very well be based upon insufficient data. This classification, therefore, is merely intended to reflect the present state of the literature with respect thereto, and is not necessarily final nor conclusive.

In substantially all the recorded cases the observers directly or indirectly assert the absence of arsenic and mineral poisons in the dves subjected to physiological test, but the kind of other impurity, if any, is not stated.

For the purpose of a comprehensive survey of the literature and legal enactments, the following tabulation is presented:

Condition of the United States color market in the summer of 1907.

 $[x=not\ on\ market.$  Figures indicate number of dealers offering sample. Cross lines separate the several groups.]

Green Table number.	Unfa- vor- able.	Favor- able.	Con- tradic- tory.	Green Table number.	Unfa- vor- able.	Favor- able.	Con- tradic- tory.	Green Table number.	Unfa- vor- able.	Favor- able.	Con- tradic- tory.
1 2 3	x x x			164 166 169	х	x 1		521 527		x x	
14 5 6	X	10 x		188 197 201	2		1 4	530 532	x x		
8			5	240 269		1	1	563			x
9 11 13			1 2 6	277 287	х		1	572 574 576	x x	x	
14 15 16			x x	394 398		х	2	584 593		x	1
17 18 28		x	2	399	3	x		599 600 601		x x	1
41 43 55			x x 2	427 428			2 3	602 614	x x		
65 70 78	x	2	x	433 434 435	1	1 4		620 639	x x		
84 85 86		2	2 8	448 450 451			4 X 5	649 650 651	X		2
87 88			x x	457 459	x		x	654	x x		
89 92		1 x		462 467		x <sup>2</sup>		659	х		
93 94	6	x		477 478	x	x		667	1.		
95 97 102	1	х	2	479 480 483	x		1 x	670 675	x x		
103 105		6	-,	488, 490	х			689 692		3	х
106		7	5	502 504 512	2	3	5	Totals 106	33	32	41
138 160 163			X X X	516 517 520	1	5 2		2 50	28	<sup>2</sup> 16	<sup>2</sup> 26

<sup>&</sup>lt;sup>1</sup> Italicized figures are colors in the permitted list, Food Inspection Decision 76. <sup>2</sup> On United States market in 1907.

This table shows that of the 106 coal-tar dyes examined physiologically only 50 were on the United States market; further, out of 33 "unfavorable" dyes 8, or one-fourth, were on the United States market; out of 32 "favorable" dyes 16, or one-half, were on the United States market, and finally that out of 41 "contradictory" dyes 26, or very nearly two-thirds, were on the United States market in the summer of 1907.

Assuming this classification to be substantially and essentially fair, the only Green Table numbers which are of interest for the present discussion are those classified under "favorable," because any color positively injurious or of doubtful character is considered as being properly excluded from use in food products. The Green numbers classified as "favorable" are 32 in number, as follows: 4; 5; 28; 65; 85; 89; 92; 93; 102; 103; 105; 107; 166; 169; 240; 394; 399; 433; 435; 462; 467; 477; 512; 517; 520; 521; 527; 576; 593; 599; 600; 692.

Of these 32, 16 were on the United States market in the summer of 1907 and their composition disclosed; they are presented in the following table, together with the number of dealers, out of a possible 12, offering them for sale.

Distribution of "favoral	le" colors on the	e American me	arket in 1907.
--------------------------	-------------------	---------------	----------------

Green	Number	Green	Number	Green	Number	Green	Number
Table	of dealers	Table	of dealers	Table	of dealers	Table	of dealers
number.	handling.	number.	handling.	number.	handling.	number.	handling.
65 85 89	10 2 2 2 1	103 105 107 169	6 1 7 1	240 433 435 462	1 1 4 2	512 617 520 692	. 3 5 2 3

From among these 16, six of the seven permitted colors of Food Inspection Decision No. 76 were selected. The process of selection and of elimination is described on page 166.

# CLASSIFICATION ACCORDING TO CHEMICAL COMPOSITION AND SUITABILITY.

In the following table the chemical composition of the substances corresponding to the Green Table numbers is given, and their classification according as the literature is regarded as being (1) unfavorable, (2) favorable, or (3) contradictory in regard to the color. The colors are also arranged in the groups to which they belong chemically, so that this table shows: (1) The number of groups reported on in the literature, (2) the number and composition of members of each group so reported on, and (3) the interpretation here placed upon such reports in literature. This table is given in the expectation that it will be of use to chemists and physiologists.

 $Opinions\ as\ to\ suitability,\ classified\ according\ to\ groups\ and\ chemical\ composition.$ 

(The chemical nomenclature is that of the Green Tables; "a" is used for alpha and "b" for beta.)

(The offention nomencature is that of the offent rands, as is used to arpha and is not seem,)						
Unfavorable.	Favorable.	Contradictory.				
NITRO COLORS.  1. Symmetrical trinitrophenol. 2. Dinitro o and p-cresol. 3. Dinitro-a-naphthol. 6. Hexanitro-diphenylamin. MONOAZO COLORS.	4. Dinitro-a-naphthol-b-mono-sulphonic acid. 5. Dinitro-a-naphthol-a- mono-sulphonic acid.					
<ul> <li>78. Methyl-benzenyl-amido-thio-xylenol-azo-a-naphthol disulphonic acid.</li> <li>94. Benzene-azo-pyrazalone-carboxy-disulphonic acid.</li> <li>97. Sulpho-o-toluene-azo-b-naphthol.</li> </ul>	<ol> <li>p-nitro-benzene-azo-a - naphthylamin p-sulphonic acid.</li> <li>a-naphthalene-azo - b - naphthol-disulphonic acid.</li> <li>p-sulphobenze e n e - a z o - a - naphthol.</li> <li>p - sulphobenzene - azo - diphenylamin-sul phonic acid.</li> <li>Diphenylamin yellow with nitro-diphenylamin.</li> </ol>	8. Amido - azo - benzene-di- and monosulphonic acid.  9. Amido - azo - toluene - disulphonic acid.  11. Benzene-azo-b-naphthol.  13. Benzene-azo-b-naphthol - b-sulphonic acid.  14. Benzene-azo-b-naphthol disulphonic acid G.				

Opinions as to suitability, classified according to groups and chemical composition—Con.

Unfavorable.	Favorable.	Contradictory.
MONOAZO COLORS—continued.	93. p-sulphobenzene-azo - dioxynaphthalene sulp h o n i c acid.  102. p-sulpho-naphthalene - azo - b-anphthol.  103. p-sulpho-naphthalene-azo-anaphthol-p-sulphonicacid.  105. p-sulpho-naphthalene - azo - b-naphthol m o n o s u l-phonic acid.  107. p-sulpho-naphthalene - azo - b-naphthol-disulphonic acid.	15. Benzene-azo-b-naphthol disulphonic acid R. 16. Dimethyl-amido - azo - benzene. 17. Hydrochlorid of diamido-azo-benzene. 18. Hydrochlorid of benzene-azo-m-tolylene-diamin. 19. Hydrochlorid of toluene-azo-m-tolylene-diamin. 19. Toluene-azo-b-naphthol-sulphonic acid. 19. Diehloro-phenol-azo-b-naphthol-disulphonic acid. 20. Diehloro-phenol-azo-b-naphthol. 21. P-sulphobenzene-azo-resorcinol. 22. P-sulphobenzene-azo-dimethylanlin. 23. P-sulphobenzene-azo-diphenylamin. 24. P-sulphobenzene-azo-diphenylamin. 25. m-sulphobenzene-azo-diphenylamin. 26. P-sulphobenzene - azo-diphenylamin. 27. P-sulphobenzene - azo-diphenylamin. 28. P-sulphonaphthalene - azo-b-naphthol-disulphonic acid.
DISAZO COLORS.  164. Sulphobenzene-azo-sul p h obenzene-azo-b-naphtholsulphonic acid.  201. Hydrochlorid of toluene disazo-m-tolylene-diamin.  277. Ditolyl-disazo-binaphthion ic acid.	<ul> <li>166. Sulphobenzene - azo-sulphobenzene - azo - p - tolyl - b - naphthylamin.</li> <li>169. Sulphotoluene-azo - toluene-azo - b - naphthol - a - sulphonic acid.</li> <li>240. Diphenyl - disazo - binaph - thionic acid.</li> </ul>	<ul> <li>138. Bisulphobenzene - disazo - anaphthol.</li> <li>160. Sulphobenzene-azo-benzene - azo - b - naphthol - monosulphonic acid.</li> <li>163. Sulphobenzene - azo - sulphobenzene-azo-b-naphthol.</li> <li>188. Disulpho-b-naphthalene-azo-a-naphthalene-azo-b-naphthol.</li> <li>197. Hydrochlorid of benzene-disazo-phenylene-diamin.</li> <li>269. Ditolyl - disazo - bi - salicylic acid.</li> <li>287. Ditolyl-disazo-bi-a-naphthol-psulphonic acid.</li> </ul>
NITROSO COLORS.	394. Dinitroso-resorcinol.	398. Nitroso-b-naphthol-b-mono- sulphonic acid. °
STILBENE COLORS.	399. Azoxy-stilbene-di-sulphonic acid.	
DIPHENYLMETHANE COLORS.  425. Hydrochlorid of imido-tetramethyl-diamido-diphenylmethane.  TRIPHENYLMETHANE COLORS.  434. Dimethyl-dibenzyl-diamidotriphenyl-carbinol-tris ulphonic acid.  459. Chlorid of heptamethyl-rosanilin chlorid.  478. Triphenyl-pararosanilin diand trisulphonic acid.  479. Triphenyl-pararosanilin-trisulphonic acid.  488. 490. Hydrochlorid of phenyl-tetra-(penta) methyltriamido-diphenyl-a-naphthyl carbinol.	<ul> <li>433. Diethyl-dibenzyl-diamidotriphenyl carbinol-disulphonic acid.</li> <li>435. Diethyl-dibenzyl-diamidotriphenyl-carbinol-tris ulphonic acid.</li> <li>462. Trisulphonic acid of rosanilin and pararosanilin.</li> <li>477. Triphenyl-rosanilin-monosulphonic acid and triphenyl-pararosanilin acid.</li> <li>467. Disulphonic acid of dimethyl dibenzyl diethyl triamidotriphenyl carbinol.</li> </ul>	427. Chlorid of tetra-methyl di-pamido-triphenyl-carbinol. 428. Sulphate or chlorid of tetra-ethyl-diamido-triphenyl-carbinol. 448. Hydrochlorid or acetate of pararosanilin and rosaninin. 450. Hydrochlorids or acetates of mono-di or trimethyl (or ethyl) rosanilins and pararosanilins. 451. Hydrochlorid of penta- and hexamethyl-pararosanilin. 457. Hydrochlorid, sulphate or acetate of triphenyl rosanilin and triphenyl pararosanilin and triphenyl-pararosanilin sulphonic acids. 480. Triphenyl-rosanilin and triphenyl-para-rosanilin sulphonic acids. 483. Aurin carbinol, oxydized aurin, methyl aurin, and pseudo rosolic acid.

Opinions as to suitability, classified according to groups and chemical composition—Con.

Favorable.	Contradictory.
512. Tetrabromofluorescein.	504. Hydrochlorid of diethyl-mamido-phenol-phthalein.
517. Tetraiodofluorescein. 520. Tetraiododichlorofluorescein. 521. Tetrabromotetrachlorofluorescein.	amuo-phenoi-phenaiem.
pound of cerulein.	
	563. Dioxy-anthraquinone-b-qui-
1 576. New Gray (composition un- known).	
trianilido-, and tetraani- lido - phenyl - phenazo - nium chlorids.	584. Diamidophenyl and tolyl- tolazonium chlorids. 601. Sulphonated indulins.
600. Indulins and nuoringins.	
	650. Chlorid of tetramethyl-di amido-phenazthionium.
-	
	ø
VVI	
692. Indigotin disulphonic acid.	689. Indigotin.
	517. Tetraiodofluorescein. 520. Tetraiododichlorofluorescein. 521. Tetrabromotetrachloro fluorescein. 527. Sodium bisulphite compound of cœrulein.  64. 65. 66. New Gray (composition unknown).  599. Mixtures of dianilido-amidotrianilido-, and tetraanilido-, and tetraanilido-, not

### PHYSIOLOGICAL ACTION OF COAL-TAR DYES.

### SUMMARY OF SYMPTOMS.

A rough summary of the symptoms noted or positively determined to be absent, the number of the deaths produced, and the number of cases in which nothing abnormal was noticed may serve as a convenient guide in considering the detailed statements hereinafter given relative to all the symptoms, clinical data, legislative and other publications, or permissions.

In the following tables are brought together most, if not all, of the recorded observations with respect to the humans and other animals upon which the physiological action of coal-tar dyes has been studied as well as the results of autopsies when recorded. The columns headed "Unfavorable," "Favorable," and "Contradictory" have the same significance as in the preceding table; italicized numbers are those of the permitted colors of Food Inspection Decision No. 76. Asterisked numbers indicate that the dye was administered hypodermically.

## EXPERIMENTS ON DOGS.

Table, I.—Observations on dogs.

(Reference to Green Table numbers.)

Symptoms.	Unfavorable.	Favorable.	Contradictory.
Deaths	1; 2; 3; 488 or 490; 574; 639 (2); 649.		16(2);55;70*;86;87;95(2); 188*; 428 (2); 451 (2);
Autopsies	3; 488 or 490; 572; 574; 620; 639; 649.	103; 105; 399; 467; 477; 593; 692.	480*; 601; 650. 16; 55; 70*; 87 (2); 45 (2); 457; 480 (2); 601; 650.
Stained conjunctivæ	1		398. 398.
Respiration difficulties Temperature normal	2: 620: 649	467	451; 601.
Temperature low	488 or 490; 3		457. 451.
General depression. Weakness. Loss of weight.	1; 3; 97; 649; 659		650. 16; 483.
Emaciation Paralysis of heart	488 or 490; 639		138; 451.
Loss of appetite	2; 3; 97; 425; 530; 649; 659; 667; 670.	240; 394	16; 87; 138; 197; 428; 451; 483; 650.
Aversion to food	649; 3	4; 5; 28; 92; 93; 103; 105;	451. 8; 9; 11; 13; 14; 16; 55; 84;
	434; 459; 478; 479; 502; 516; 530; 532; 614; 620; 651; 659; 667; 670.	166; 240; 394; 399; 433; 521; 527; 576; 599.	86; 87; 88; 95; 138; 197; 269; 287; 398; 428; 448; 483; 504; 601; 650.
Colorless urine	400 400- 640	240	
Albuminuria	1; 3; 78; 94; 97; 425; 434; 459; 479; 516; 580; 651; 659; 667.	467; 477 4; 5; 28; 166; 240; 394	451; 480; 601. 8; 9; 11; 17, 18, or 41; 70; 86; 88; 95; 188; 197; 269; 287.
Albuminuria doubtful	78; 478	5240	138. 14; 88; 269; 287.
Bloody urine		210	451.
Kidney irritation	1		86. 650.
Thirst	94; 425; 488 or 490; 620	103; 105; 399; 477	87; 88; 398; 480; 601; 650.
Thin stool. Diarrhea	1; 3; 94; 97; 479; 425; 516;	527	11; 16; 84; 86; 87; 138; 269;
Softened feces			428; 483; 504; 584; 650. 95. 650.

Table I.—Observations on dogs—Continued.

Symptoms.	Unfavorable.	Favorable.	Contradictory.
Pus in stool			650.
Vomiting		399; 433; 467; 576; 692	11; 16; 86; 87; 95; 197; 269; 428; 448; 451; 483; 650.
Retching Salivation Anemia	675 488 or 490; 639; 649; 675	28	584.
Stupor			601. 451.
Catarrh of eyes and nose Cramps Convulsions	1; 2		601.
Tremors	649		87.
Destroys coloring matter of blood.	1; 2		
Loss of sight. Loss of hearing. Internal disorders.			87; 601. -601. -650.

The following table discloses the results of the autopsies made and reported on dogs.

Table II.—Autopsies on dogs.

	1	1	
Symptoms.	Unfavorable.	Favorable.	Contradictory.
No change in internal			87.
organs. Nothing abnormal		105; 399; 467; 477; 593	87 III: 457.
No change except whole interior colored red.		103	S. 222, 2317
LIVER.			
Tatty degeneration	572; 620; 639		601; 650.
Blood-poor Pale			451 (2); 480 II*; 601. 451 (2).
wollennflamed			650.
Blood-filled			000.
KIDNEYS.			
Congested	3; 639; 649		451 (2); 480* (II).
filled with decomposed blood corpuscles.	639		451 (2).
colored			480 (1); 601; 650.
oftBlood-poor	488 or 490		480* II. 601.
Blood-filled	574		
Nhickened	620; 639	09%	650.
nflamed			650.
STOMACH.			
olored			650.
atarrhnflamed mucous mem-	488 or 490		650.
brane. unctured mucous mem-			601.
brane.			001.
ontracted and filled with colored mucous.	639		
LUNGS.			
Filled with blood	574		601.
HEART.	Man 4		
wollen Paralyzed	574. 574.		16 (2); 87 I; 428; 87 I

Symptoms.	Unfavorable.	Favorable.	Contradictory.
intestines. Colored Catarrh	488 or 490		650.
			650. 650.
kin			650.
Heart sac			650. 650.
Esophagus	488 or 490		650. 480* IT.
All organs swollen			480* II.
Peritonitis	3 (II)		70*.
Fat all disappeared Flabby muscles	639		

Table II.—Autopsies on dogs—Continued.

In this connection it may be of interest to note that out of 16 dyes producing death when administered through the mouth to dogs, 7 were on the United States market in the summer of 1907. Their Green Table numbers are as follows, the numbers in parentheses indicating the number of makers or importers, out of a possible 12: offering them: 55 (2); 86 (8); 95 (2); 428 (3); 451 (5); 601 (1); 650 (2).

The following table gives the Green Table numbers of coal-tar dyes which on administration to dogs positively did not in certain specific cases produce the particular disturbances recited, although the case of the several colors as a whole is regarded in the literature as "favorable," "unfavorable," or "contradictory," as stated.

Table III.—Observations on dogs showing definite negative results in specific cases grouped under the general verdict of the literature as a whole.

Symptoms.	Unfavorable.	Favorable.	Contradictory.
Albuminuria	6; 97; 277; 434; 532; 614; 651; 659.	4: 105: 462	95: 504
Loss of weight		692	86.

That is, when these dyes were tried on dogs the observers reported in certain specified cases, positively and definitely, the absence of any of the symptoms named.

It is stated of the following colors that they produced no bad effects in certain specified cases: 4; 9; 65; 86; 105; 188; 197; 240; 457; 572; 593. No bad effect except colored urine: 13, 95. No bad effect except albuminuria: 88, 287. No bad effect except loss of weight: 12.5 per cent (17, 18, or 41).

It is therefore true of all of these coal-tar dyes that there are conditions under which they have been observed not to produce the bad

effects as above set forth; but this tabulation must not be taken to mean that these dyes can not, under any conditions whatever, produce untoward results; the reverse is true in most cases.

### EXPERIMENTS ON HUMAN BEINGS.

The Green Table numbers of those colors concerning which experimental data are available on humans are as follows: 1; 2; 3; 4; 6; 9; (17, 18, 41); 55; 65; 85; 86; 95; 102; 103; 105; 106; 107; 197; 427; 428; 448; 462; 532; 602; 650.

The symptoms produced are classified in the following list:

## Internally administered.

Not poisonous	Bad taste in mouth
9; 55; 65; 85; 95; 102;	Restlessness
103; 105; 106; 107; 448; 462	Rush of blood to head
Poor general condition 86; 650	Vertigo 86; 650
Fever	Headache
Loss of appetite	Delirium
Vomiting 3; 532; 650	Twitching of muscles 650
Intestinal irritant	Yellow coloration of skin
Diarrhea 532; 650	Yellow-colored mucous membrane. 3
Bladder irritant	Food colored with it made a family
Colored urine 86; 95; 650	sick 3
Albuminuria	Adults withstand 1
Increased micturition 650	Children and weak adults do not
Irritant 532	withstand
Inflammation	Deaths 2; 3
Dryness of throat	Autopsy <sup>1</sup> 3

It should be noted that of the 13 dyes here classed as not poisonous to humans all but No. 102 were on the United States market in the summer of 1907, as is shown in the following table:

Number of dealers offering these nonpoisonous colors in 1907.

Green	Sources offering same.	Green	Sources	Green	Sources	Green	Sources
Table		Table	offering	Table	offering	Table	offering
numbers.		numbers.	same.	numbers.	same.	numbers.	same.
1 4	10	65	2	103	6	107	7
9	1	1 85	2	105	1	448	4
55	2	95	2	106	5	462	2

<sup>&</sup>lt;sup>1</sup> On permitted list, Food Inspection Decision 76.

Symptoms produced by external application of certain colors (Green Table numbers).

Burning 2; 427; 428	Dermatitis 4
Itching	Eczema (17, 18, or 41); 197; 602
Blisters	
Swelling	

It should be noted that No. 86 has been tried on humans and has been found not to produce diarrhea or vomiting.

<sup>&</sup>lt;sup>1</sup> Hemorrhagic gastritis.

G. T. 3 has apparently killed a human at 60 mg per kilo and the autopsy disclosed hemorrhagic gastritis. G. T. 448 has been suggested as a possible remedy for Bright's disease. It should also be borne in mind that adults can stand G. T. 1 in doses of 540 to 900 mg daily for a long time, whereas children and weak adults stand that substance only poorly.

### EXPERIMENTS ON SMALL ANIMALS.

Results of experimenting on rabbits with 10 coal-tar dyes whose Green Table numbers are 1, 2, 86, 89, 107, 427, 448, 504, 517, and 563 have been tabulated as follows:

Death	563*	Paralysis
Diarrhea	1	Cramps
Colored urine	86	No harm produced. 89; 107; 448; 504; 517
Softened feces	86	

In the case of the numbers marked with an asterisk the color was administered hypodermically. In this connection reference should also be had to the paper of Penzoldt abstracted in Section VIII, page 55.

The four coal-tar dyes 55, 103, 425, and 480 have been tested on guinea pigs and no disturbance was noticed in all, but in the case of 103 occasional thirstiness was observed. No. 448 has been fed to hens without damage, and No. 2 has been recommended as an insecticide, a fungicide, and a mouse poison.

## GENERAL STATEMENTS.

The following statements may be of interest before the detailed compilation is read:

1. O. Buss (Forschungsber. über Lebensmittel, 1896, vol. 2, pp. 163-197, 237), in a paper entitled "Contributions to the Spectrum Analysis of some Toxic and Pharmacognostically Important Coloring Matters, with Special Consideration to the Ultra-Violet," cites as poisonous the following (Green Table numbers follow in parentheses wherever connection could be satisfactorily established):

Picric Acid	(1)	Metanil Yellow(95)
Dinitro o- and p-Cresol	(2)	Corallin(483)
Martius Yellow	(3)	Safranin(584)
Aurantia	(6)	Methylene Blue(650)
Fast Yellow	(8)	Iodin Green (459)
Orange II	(86)	Alizarin Blue S (563)
As nonpoisonous:		
Naphthol Yellow	(4)	Naphthol Green (398)
		Malachite Green (427, 428)
		Dahlia(450, 451)
Anilin Blue	(457)	
As doubtful:		

## On the following Buss is noncommittal:

Auramin	(425)	Magenta	(448)
Biebrich Scarlet	(163)	Aurin	1(483)
Water blue	(480)	Acid Green	(435)

Naphthol Black of various brands, which seemed to be mixtures.

# 2. It has been pointed out that the following nine colors are harmless:

Naphthol Yellow.       (4)       W         Naphthol Brown.       (?)       M         Chinolin Yellow.       (667)       A         Pyrotin RRO.       (115)       F         Acid Green.       (434, 435)       F	Milling Red
-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------	-------------

Most of these have not been examined experimentally, but scientific studies have been made of the poisonous qualities of Azo-Blue and Naphthol Yellow. (Zts. angew. Chemie, 1896, p. 24.)

## 3. Chlopin in his monograph (see p. 75) says:

On the basis of my personal experience I consider the testing of the action of coaltar dyes on man not permissible, since such experiments may induce in the subjects of experiment more or less serious symptoms of poisoning, for which in some cases there are no antidotes at our disposal. To such accidents, in my opinion, only the experimenter himself may subject himself, because he knows what he is doing. Preliminary tests of dyes on dogs and other animals afford no guaranty of escape from disagreeable accidents which may take place in the testing of the dye on man. (Page 111.)

These data and calculations convincingly prove how erroneous the current opinion is that for the coloring of food substances and beverages only exceedingly small, almost unweighable, quantities of dyes are used. (*Page 113*.)

On page 221 et seq., the following general discussion of this subject is found:

THE MANNER OF ACTION OF POISONOUS DYES ON THE ANIMAL ORGANISM.

The mechanism and the chemistry of the action of the artificial dyes of the aromatic series on the animal organism remains to the present day, with few exceptions, exceedingly slightly and superficially studied. The same can be said also concerning the pathological and anatomical changes which are induced by these dyes.

More than the others there have been studied in the toxicological respect the dyes belonging to the Nitro group; Picric Acid; Martius Yellow (Dinitro-naphthol potassium), and Saffron substitute (Dinitrocresol potassium.)

According to Kobert, these dyes belong to the poisons acting on the blood. According to the same authority, Methylene Blue, which belongs to the other chemical group of Thiazins, acts similarly.

In the fundamental works on the sanitary investigations for the dyes, by Cazeneuve and Lépine, by Weyl, and by Santori, we find almost no material relating to the explanation of the manner of the action of the dyes. These investigators limiting themselves to a very cursory description of the symptoms of poisoning, and reciting in most general terms the results of autopsy, not even indicating the cause of death. Such, for instance, are the reports of autopsies made by T. Weyl and some other investigators, as quoted above.

We may expect that more detailed investigations in this respect will be made at the proper time by pharmacologists, since study of the mechanism and chemistry of the action of the poisonous substances on the animal organism is their province; for the hygienist it is quite sufficient merely to establish the fact that a given substance is poisonous or harmful, and he need not go any further. For this reason, in those cases in which I desired to clear up the causes of death of the animals in my experiments, and to record pathological and anatomical changes (although by the terms of the regulations governing this competition, a close study of the action of the dyes, and the ascertainment of the mechanism and chemistry of their action was not required), I called in a person more competent than myself on these questions.

Not counting the duplicates we made five autopsies all told. In all these cases death resulted from paralysis of the heart. The pathological and anatomical changes in all cases, except one, did not present anything specific, and finally reduced themselves to a feebly expressed turbid swelling of the heart and of the liver, a rush of blood to the

stomach, and a congestion of the internal organs.

The exception was the autopsy of a dog, which died from Methyl Orange; this dog died with the symptoms of paralysis of a cerebro-spinal nature. This experiment was made twice, and the autopsies of both animals showed hyperæmia in the lowest part of the spinal column, on the border of the anterior and the lateral columns.

As to the symptoms of poisoning not resulting in acute death, here most frequently was observed vomiting, diarrhea, and albumen in the urine, showing disturbance of

the functions of the digestive tract, and an affection of the kidneys.

A highly typical picture of poisoning is presented by the sulphid Vidal dyes. They cause rapid, almost instantaneous, deafening of the animal, whereupon the animal falls on one side in convulsions and lies, not moving its body, but convulsively and rapidly twitching its anterior limbs during several minutes. The tongue hangs out of the mouth, a strong secretion of saliva is noted, then vomiting begins, and the dog gradually begins to revive; with difficulty he arises on his front feet and sits down, not being able yet to stand on his posterior extremity, which is in a state of paresis. After a few hours the dog becomes normal. The symptoms of poisoning, just described, are exceedingly similar to the supposed "apoplectic form" of poisoning by hydrogen sulphid, which had been studied on animals by K. Lehmann, and which was observed in persons who inhaled air containing a few per cent of this gas. Air containing 0.1 to 0.3 per cent of hydrogen sulphid kills cats and dogs in 10 minutes.

In our experiments in which were introduced substances containing sodium sulphid, the poisoning must have been caused by hydrogen sulphid which was liberated from the dye by the acid of the gastric juice, and which could cause poisoning also through the stomach and through the respiratory apparatus.

Fortunately Vidal dyes, owing to their repulsive odor, will scarcely find a wide

application in coloring food and beverages.

Some Reflections Regarding Further Investigations of Dyes from a Sanitary Standpoint.

The present investigation, as well as all the investigations of the action of dyes on animals by previous investigators, had for its object the solution of the question in what number there exists among the dyes of the aromatic series dyes which possess poisonous, or more or less pronounced harmful properties (answering essentially the sanitary toxicological question). From the practical point of view such investigations presented and do present the most important interest, inasmuch as they afford a possibility of protecting the public from the use of obviously poisonous and harmful substances, but by such investigations questions of exceedingly sanitary importance are not answered, namely:

1. Ought we to consider as quite harmless those dyes which do not induce pronounced symptoms of poisoning and which are designated herein by the term non-poisonous?

2. Is the usual answer which is given to the hygienist by the defenders of the unrestricted use of the coal-tar dyes for coloring food products and beverages, namely, that in practice the dyes are introduced into the human organism in so small quantities that their properties can be neglected, justified?

To both questions, besides the facts and considerations which I gave above in my

investigations, we may reply experimentally in two ways:

(a) By prolonged investigations continued over a period of years of the action of very minute quantities on the animal organism, which has so far, owing to the incon-

veniences of such long experiments, not been done by anybody; and

(b) By investigation of the action of small doses of dye on some physiological functions, and first of all on the activity of the digestive organs, which is first of all disturbed upon the introduction of dyes into foods and beverages. The solution of the last question can be best promoted, in my opinion, by experiments made on dogs and by exploratory examination of the body according to the method of Prof. Pawlow. Unfortunately, experiments such as these, owing to the difficulty of the Heidenhain-Pawlow operation, are inaccessible to the majority of investigators.

As a very useful substitute of such investigations may serve observations on the action of dyes on the activity of the digestive juices outside the body of animals.

On my proposition Dr. A. E. Winogradow began in my laboratory experiments on the action of small doses of dyes of the aromatic series on the digestion in vitro. Dr. Winogradow so far examined 25 coal-tar dyes in this respect according to the method of Metta and convincingly proved that in insignificant doses coal-tar dyes (from one-half to 4 milligrams) entirely stopped the digestion of albumen by artificial gastric juice. It was found that the capacity to depress the digestion is possessed not only by poisonous dyes, but also by dyes which proved in my experiments on animals nonpoisonous.

It is quite possible, therefore, that an admixture of coal-tar dyes will exert an unfavorable influence on the digestion and assimilation of food prepared from products colored by them. Experimental proof of the last supposition can be given only by experiments on the influence of dyes on the metabolism of substances in animals and man.

### COMPLETE DETAILED STATEMENT OF ALL COMBINED DATA.

#### ABBREVIATIONS OF AUTHORITIES CITED.

The data hereinafter given is brought together as nearly as possible under the Green Table numbers to which it is pertinent. It is believed that the literature has been quite thoroughly searched, and that nothing of substantial importance has escaped recording in this compilation; certainly whatever may have escaped can hardly serve to change the general conclusion to which this compilation leads.

In order to avoid repetition in the following tabulation, "Weyl" is to be understood as referring to the book entitled "The Coal Tar Colors, with Especial Reference to their Injurious Qualities, etc.," by Theodor Weyl, translated by Leffmann and published in Philadelphia, Pa., in 1892.

"Lieber" refers to the book entitled "The Use of Coal Tar Colors in Food Products," by Hugo Lieber, published in New York in 1904.

"Fraenkel" refers to the book entitled "Arzneimittel Synthese," by Dr. Sigmund Fraenkel, published in Berlin in 1906.

"Confectioners List" refers to the Official Circular from the Executive Committee of the National Confectioners' Association of the

United States entitled "Colors in Confectionery" and reprinted, in part, in the book entitled "Food Inspection and Analysis," by Albert E. Leach, published in New York in 1906, pages 630-634.

"Resolutions of Swiss Analytical Chemists" refers to these resolutions as published in Zeitschrift für Nahrungsmittel Untersuchung

und Hygiene, 1891, page 293.

"Schacherl" refers to Schacherl's publication entitled "Die Zulaessigkeit Kuenstlicher Farbstoffe zum Färben von Lebensmitteln," published in Vol. III, pages 1041–1048, of the Report of the Fifth International Congress of Applied Chemistry held in Berlin June 2 to 8, 1903.

"Chlopin" refers to Chlopin's monograph published in Russian and entitled "Coal Tar Dyes. Classification, properties, and action of artificial dyes on the animal organism, etc.," published at Dorpat in 1903, or to the abstract of Chlopin's paper printed at page 169–172 of Vol. IV of the Report of the Fifth International Congress of Applied Chemistry held in Berlin, 1903.

"Canton of Tessin" refers to the publications of the Tessin regulations published in 1897 in Zeitschrift für Untersuchung der Nahrungs

und Genussmittel, page 414.

Whenever possible the doses administered have been given in milligrams per kilo and grains per 100 pounds of body weight of animal. In the case of the tabulations taken from Chlopin's monograph this was not done; but in order to render such comparative data easily available factors have been placed at the head of each tabulation; for example, under G. T. 6 (1 gram=106 mg=74.2 grains), which means that each gram administered amounts to 106 mg per kilo or 74.2 grains per 100 pounds of body weight of animal; by multiplying the doses given by either of the factors the corresponding comparative information is obtained.

In addition to the 106 Green Table numbers that have been examined physiologically, there are reported the results of the physiological examination of 8 coal-tar colors not in the Green Tables, of which 3 are said to be nonpoisonous, 3 are called poisonous, 1 is called harmful, and the last is said to be "not quite harmless." These 8 dyes are not included in this compilation.

TABULATION BY GREEN TABLE NUMBERS OF PHYSIOLOGICAL AND OTHER DATA.

## G. T. 1.

Trade names.—Picric acid; carbazotic acid. Scientific name.—Symmetrical trinitrophenol. Discovered.—1771.
Shade.—Yellow. Not offered.

Nothing.

### UNFAVORABLE.

1. Prohibited by Confectioners' List.

2. Weyl (p. 30): "The injurious character of picric acid has long been known."

3. "In Germany its employment for coloring food is forbidden by the imperial enactment of 1888, on account of its poisonous character." (pp. 68-71.)

4. "The foregoing statements show that while the acid must be considered poisonous, its injurious character is far less than has generally been assumed, nevertheless, the legal prohibition of its use as a coloring matter for food or drink is just." (p. 71.)

"Erb gave a rabbit weighing 1,700 grams, 0.06 gram of potassium picrate (24.5 grains per 100 pounds) daily for 90 days; slight loss of weight and occasional

diarrhea were noted, but nothing more serious." (p. 69.)

6. A rabbit weighing 2,065 grams died at the end of 19 days, after having taken 2.52 grams of the substance, or 854 grains per 100 pounds body weight; number of

doses not stated. (p. 69.)

- 7. Weyl's experiment on a dog, weight not given: April 21–26, 0.24 gram (3.7 grains) sodium picrate daily; April 28–May 9, 0.36 gram (5.5 grains) daily; total, 5.76 grams (8.9 grains) sodium picrate; no serious disturbance; May 13, 1.2 grams (18½ grains) sodium picrate at one dose; weakness marked, diarrhea and dyspnea next day; May 14, 0.6 gram (9¼ grains) caused vomiting; evening of same day, 0.36 gram (5.6 grains) given; May 15, animal lively; 0.24 gram (3.7 grains) again given, and on evening of same day 0.72 gram (11.2 grains); May 16, marked weakness of animal, and 0.16 gram (2.5 grains) given, causing vomiting; May 17, 0.17 gram (2.6 grains) given; May 18 and 19, animal definitely recovered, and aside from strong yellow tinge of the conjunctiva and skin, no abnormal conditions manifest. Animal died May 20 after receiving 1.32 grams (20.4 grains) potassium picrate. Weyl concludes, therefore, that dogs are resistant to this substance, notwithstanding the prostration and the blood disorganization.
- 8. Weyl summarizes the effect on humans from therapeutic and poisoning cases to the effect that daily doses of from 0.54 to 0.90 gram (8.3 to 13.8 grains) of potassium picrate are easily borne by healthy adults for a considerable time; children and weak adults bear picric acid badly. (p. 70.)

9. "Picric acid \* \* \* is poisonous \* \* \*." (p. 96.)

10. Fraenkel (p. 572): "On the other hand, this substance is not usable for internal administration on account of its decomposing the red blood corpuscles, and of its energetic cramp production, as well as on account of its disturbance of the kidneys, and the ultimate paralysis of the respiratory centers; nevertheless, picric acid is not to be considered a violent poison \* \* \*."

11. Schacherl (p. 1044): "Picric acid \* \* \* (is), according to numerous statements in the literature, poisonous even in small doses, and (is) therefore un-

qualifiedly to be declared as unpermissible."

12. Lieber (p. 16), where it is stated to be forbidden by the German law, and is also otherwise substantially the same as Weyl above quoted.

13. Resolutions of the Society of Swiss Analytical Chemists, September, 1891: "The following are to be regarded as coloring matters harmful to health: \* \* \* picric acid \* \* \* \*."

14. Prohibited by the Belgian law of June 17, 1891.

15. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Picric acid is poisonous. Rabbits can stand daily 10 milligrams of a green containing picric acid, but not 20 milligrams. Their death is accompanied by paralysis."

16. Buss lists it as poisonous.

## G. T. 2.

Trade names.—Victoria Yellow; Victoria Orange; Golden Yellow; Saffron Substitute; Anilin Orange; Di-nitro-Cresol.

Scientific name.—Di-nitro-o-and-p-cresol.

Shade.—Yellow. Not offered.

Discovered.—1869.

Used for coloring butter, liqueurs, etc.

FAVORABLE.

Nothing.

UNFAVORABLE.

- 1. Prohibited by Confectioners' List.
- 2. Fraenkel (p. 572): "On the other hand dinitro-cresol is much more intensely poisonous (than picric acid), which is probably caused by its greater solubility in water."
- 3. Schacherl (p. 1044): \* \* \* Dinitro-cresol[is], according to numerous statements in the literature, poisonous even in small doses, and [is] therefore unqualifiedly to be declared as unpermissible."
- 4. Resolutions of the Society of Swiss Analytical Chemists, September, 1891: "The following are to be regarded as coloring matters harmful to health \* \* \* Dinitro-cresol \* \* \*."
- 5. Forbidden by the Canton of Tessin.
- Weyl (p. 31): "I have shown the same (poisonous nature) for Dinitro-cresol (Saffron Substitute). (See Zts. angew. Chem., 1888, No. 12, for confirmation of my results by Gerlach.)"
- "The reverse is the case with the poisonous dinitro-cresol (Saffron Substitute)." (p. 55.)
- 8. Weyl describes experiments with this compound. (pp. 71-85.)
- 9. Fourteen rabbits were experimented on, of which 13 died. Amounts administered in the fatal cases per 100 pounds body weight were (p. 74):

Grains.	Grains
189	175
175	168
175	175
175	168
175	168
175	175
175	

Of 12 experiments on dogs, 5 receiving the color by the mouth and 7 hypodermically, 3 cases resulted fatally; the fatal case by the mouth requiring  $38\frac{1}{2}$  grains per 100 pounds body weight; the 2 fatal cases hypodermically represented 11 and 20 grains per 100 pounds body weight, respectively, although 140, 38.5, 31.6, and 35 grains per 100 pounds body weight by the mouth were borne without fatal effect; and  $24\frac{1}{2}$ , 11.9, 9.8, and 4.9 grains per 100 pounds body weight, hypodermically, were also borne without fatal effect (p.75).

- 10. Weyl (p. 96): "\* \* \* Dinitro-cresol \* \* \* are [is] poisonous; \* \* \*"
- 11. Prohibited by the Belgian law of June 17, 1891.
- 12. (Zts. Nahr. Genussm., 1892, p. 353): Recommended as an insecticide, 1500 being sufficient for all ordinary purposes. One milligram is sufficient to kill a mouse; 2 milligrams recommended for killing mice.
- 13. Weyl (Handbuch der Hygiene): For humans, the fatal dose, when administered by the stomach, appears to be 60 milligrams per kilo body weight, or 43 grains per 100 pounds.

- 14. "The president of the Council of Oppeln forbids on April 19, 1899, the use of Saffron Surrogate for coloring food products."
- 15. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Saffron Surrogate '\* \* \*, which is used for coloring foodstuffs, is poisonous. It appears to attack the coloring matter of the blood, and produces, in the case of dogs, vomiting, cramps, and convulsions. \* \* \* Feathers colored with Saffron Surrogate cause burning and itching, and finally blisters on the hands of the women working with them; the faces were also similarly affected, and this was accompanied by loss of appetite and fever."

16. Buss lists it as poisonous.

## G. T. 3.

Trade names.—Martius Yellow; Naphthol Yellow; Naphthylene Yellow; Naphthylamin Yellow; Manchester Yellow; Golden Yellow; Saffron Yellow; Jaune d'Or; Jaune Naphthol.

Scientific name.—Dinitro-alpha-naphthol.

Discovered.—1864.

Shade.—Yellow. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

- 1. Prohibited by Confectioners' List.
- 2. Weyl (p. 31): "Cazeneuve and Lépine pointed out the poisonous nature of Martius Yellow \* \* \*."
- 3. "This body (Chamber of Commerce at Sonneberg) recommends for the preparation of children's toys three colors, the poisonous character of which I can demonstrate. These are Martius Yellow \* \* \*." (p. 34.)
- 4. "For instance, for preliminary researches, dogs and rabbits have value for chemical reasons. The conclusions derived from such experiments must be accepted with great deliberation, since it happens that rabbits will bear without injury doses which will seriously, nay, even fatally, act upon the dog, as I have already shown to be the case with Martius Yellow." (p. 56.)
- 5. Where two experiments by Cazeneuve and Lépine are referred to, in which diarrhea, vomiting, and albuminuria were produced by this substance. (pp. 85-89.)
- 6. Weyl's own experiments on 4 dogs showed weakness, vomiting, diarrhea, and albuminuria resulting from the use of this color; the amounts of color administered per kilogram of body weight were 73, 17.5, 17.5, and 11.3 milligrams, which amount to 51, 12, 12, and 8 grains, respectively, per 100 pounds of body weight. (p. 87.)
- 7. "Martius Yellow, therefore, belongs to the injurious colors. As a coloring matter for food and drink its use should be wholly prohibited." (p. 89.) 8. "\* \* \* and Martius Yellow are poisonous; \* \* \*." (p. 96.)
- 9. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): "Like Saffron Surrogate, it is poisonous. In an experiment on myself, using large doses, I noticed among others the general vellow coloration of the skin. In a poisoning resulting fatally after 5 hours with Martius Yellow, vomiting, yellow coloration of the skin and mucous membranes were observed; whereas the autopsy revealed, among other things, hemorrhagic gastritis. (Jacobson, Hosp. Tid., 1893, p. 765.)"

- 10. "Such small amounts as are used for the coloring of pastry are said to be non-poisonous. (Vitalil boll. chim. farm., 1893, p. 738.)" (p. 231.)
- 11. Cazeneuve and Lépine (Compt. rend., 1885, v. 101, pp. 1167-1169) say: I. "A dog received 71 milligrams per kilogram of body weight, or 50 grains per 100 pounds daily. On the second day diarrhea and vomiting ensued; loss of appetite except for milk. Thereafter it experienced difficulties in breathing; suffered albuminuria; its urine was colored; it died on the sixth day. The autopsy disclosed considerable congestion. II. A dog weighing 22 kilos received 400 milligrams (19 milligrams per kilo or 13 grains per 100 pounds); this caused a yellow vomit; next day it received 500 milligrams (27 milligrams per kilo or 17 grains per 100 pounds), which caused violent diarrhea, fever, thirst, disinclination for all food. The animal was killed; the autopsy showed badly congested kidneys."

12. Prohibited by the Belgian law of June 17, 1891.

13. Schacherl (p. 1044): "\* \* Martius Yellow \* \* \* [is], according to numerous statements in the literature, poisonous even in small doses, and [is] therefore unqualifiedly to be declared as unpermissible."

14. Prohibited by law in Italy. (See Lieber, p. 24.)

- 15. Fraenkel (p. 572): "This substance also shows poisonous properties, although it is less poisonous than dinitro-cresol."
- 16. Resolutions of the Society of Swiss Analytical Chemists, September, 1891: "The following are to be regarded as coloring matters harmful to health \* \* \* Martius Yellow \* \* \*."
- 17. Forbidden by the Canton of Tessin.
- DIETRICH (Zts. Nahr. Genussén., 1902, v. 5, p. 364): "A lot of groats, after eating which a family became sick, was found to be free from ordinary poisons, but had been colored with Martius Yellow."
- 19. Buss lists it as poisonous.

#### DOUBTFUL.

 WINOGRADOW (Zts. Nahrs. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 4.

Trade names.—Sulphur Yellow; Sulphonaphthol Acid Yellow; Succinic; Solid Yellow; Saffron Yellow; Jaune Acide C.; Jaune Acide; Fast Yellow; Citronin; Anilin Yellow; Acid Yellow S.

Names under which it was offered on the United States market as a food color in 1907.—Naphthol Yellow SLOZ; Naphthol Yellow S; Naphthol Yellow; Naphthol Yellow L; Yellow F Y; Lemon Yellow.

Scientific name.—Dinitro-alpha-naphthol-beta-monosulphonic acid. Discovered and patented.—1879.

Shade.—Yellow. Offered by 10 out of 12 sources.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. CAZENEUVE AND LÉPINE (Compt. rend., 1885, v. 101, pp. 1167-1169): "A dog received each day for 5 days 32 milligrams per kilogram of body weight, or 23 grains per 100 pounds; for the 10 days next succeeding it received four times that amount, that is, 133 milligrams per kilogram of body weight, or 93 grains

- 2. CAZENEUVE AND LÉPINE (Compt. rend., 1885, v. 101, pp. 1167-1169)—Continued. per 100 pounds; for the 10 days next succeeding it received daily twice the last amount, or 266 milligrams per kilogram of body weight, that is, 186 grains per 100 pounds. It received altogether in the 25 days 62½ grams, or 964 grains. There was no vomiting, no diarrhea, and no albumen in the urine at any time."
- 3. Weyl (p. 31): "\* \* \* not poisonous to human beings and dogs: Naphthol Yellow S. \* \* \*."

Weyl describes his own experiments on 3 dogs, giving them, respectively, 417, 34, and 100 milligrams per kilo body weight, or per 100 pounds 292, 24, and 70 grains, respectively. Whether the color was administered by the mouth, or injected subcutaneously, all bodily functions appeared to remain normal, and it was only in the case of repeated doses of 417 milligrams per kilogram of body weight, or 292 grains per 100 pounds, that albuminuria appeared. (pp. 89-92.)

- 4. "Only the sulphonated colors Naphthol Yellow \* \* are harmless and applicable to the coloring of food and drink." (p. 96.)
- 5. Permitted by the laws of Austria.
- 6. Permitted by the law of Italy.
- 7. Permitted by the law of France.
- 8. Schacherl (p. 1044): "\* \* \* Naphthol Yellow S \* \* \* possesses no poisonous properties."
- 9. Fraenkel (p. 572): "Naphthol Yellow S is an entirely nonpoisonous substance."
- 10. Meyer (J. Amer. Chem. Soc. 1907, v. 29, p. 900): One hundred milligrams per kilogram of body weight for the initial administration, and subsequent administrations increased geometrically. After the second administration intermittent diarrhea resulted, emphasized by increased amounts with no albumin or sugar in the urine; continued for 14 administrations; so that in 14 administrations 147.58 grams of color had been given; the initial dose is 70 grains per 100 pounds of body weight, and the average daily dose of the total administered is 394 grains per 100 pounds of body weight. Urine only slightly yellow colored after small doses, but red after larger doses.
- 11. Lieber (p. 143): A dog received 36 milligrams per kilogram of body weight, or 25.2 grains per 100 pounds once a day seven times every other day; during the whole period the dog was apparently in good condition with no bad effects from the color.
- 12. Buss lists it as nonpoisonous.
- 13. CAZENEUVE AND LÉPINE (Compt. rend., 1885, v. 101, pp. 1167-1169): Three chronic invalids received daily from 2 to 4 grams of the dye in cochets; except slight colic and diarrhea nothing abnormal.

### DOUBTFUL.

- 1. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Acid Yellow S is said to be able to produce dermatitis on frequent contact therewith."
- 2. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 5.

Trade name.—Brilliant Yellow; Naphthol Yellow S or RS.

Scientific name.—Dinitro-alpha-naphthol-alpha-monosulphonic acid.

Discovered and patented.—1884.

Shade.—Yellow. Not offered.

- 1. Permitted by Confectioners' List.
- 2. Weyl (pp. 92-94): Describes experiments on 2 dogs, in which the initial dose was 532 milligrams by the mouth, and 17 milligrams hypodermically, per kilogram of body weight, respectively, 372 and 12 grains per 100 pounds of body weight; in both cases the urine was colored, in the second case traces of albuminuria resulted. In the first case the albuminuria was doubtful.
- 3. "Brilliant Yellow is not poisonous, even in large doses, when administered by the stomach \* \* \* the albuminuria was very slight. (p. 94.)
- 4. "Only the sulphonated colors \* \* \* Brilliant Yellow, are harmless, and applicable to the coloring of food and drink." (p. 96.)
- 5. Schacherl (p. 1044): " \* \* \* Brilliant Yellow \* \* \* possesses no poisonous properties."
- 6. Fraenkel (p. 572): "For the same reason \* \* \* Brilliant Yellow \* \* \* is without effect."

## G. T. 6.

Trade names.—Aurantia; Nitrodiphenylamin; Imperial Yellow; Kaiser Yellow.

Scientific name.—Hexanitro-diphenylamin.

Discovered.—1873.

Shade.—Yellow. Not offered.

FAVORABLE.

Nothing.

### UNFAVORABLE.

- 1. Weyl (p. 96): " \* \* \* Aurantia suspicious."
- 2. Schacherl (p. 1044): " \* \* \* Aurantia [is] according to numerous statements in the literature poisonous even in small doses, and [is] therefore unqualifiedly to be declared as unpermissible."
- Chlopin (p. 116) as results of his experiments considers it injurious. The experimental data are as follows:

### Experimental data by Chlopin.

#### [1 gram=106 mg=74.2 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 12 13 14 15 18 19 20 21 22–26 Total.	Grams.  1 2 3 3 3 3 3 3 3 1 18	Kilos. 9. 4	cc. 450 425 400 390 410 400	Before experiment urine and dog normal. Vomited several times; urine orange, and no albumen. Repeated vomiting; urine dark brown, acid; no albumen; dog eats. No vomiting; urine almost black; acid; no albumen. No vomiting; urine almost black; acid; no albumen. Vomiting; urine almost black; acid; no albumen. No vomiting; urine almost black, acid; no albumen. Same; urine chocolate brown, acid; no albumen; general condition normal. Gradually color of urine becomes normal; in every other respect dog is well.

- 4. Fraenkel (p. 573): "The coloring matter called Aurantia, which is a salt of hexanitro-diphenylamin, appears to be poisonous on account of the nitro groups, which is on the other hand denied by a few observers."
- 5. Resolutions of the Swiss Analytical Chemists, September, 1891: "The following are to be regarded as coloring matters harmful to health \* \* \* Aurantia \* \* \* \* "
- 6. Forbidden by the Canton of Tessin.
- 7. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Aurantia has a poisonous action. After wearing gloves for 8 hours made with so-called dogskin, which were colored with Aurantia, a man suffered confluent blisters, accompanied by itching. The workmen with this material get blisters on the face and on the hands. Perspiration increases the tendency to such blisters."
- 8. Buss lists it as poisonous.

## G. T. 8.

Trade names.—Acid Yellow; Fast Yellow G; Acid Yellow G; Fast Yellow; Fast Yellow extra; Jaune Acide; New Yellow L.

Names under which it was offered on the United States market as a food color in 1907.—Fast Yellow Y; Fast Yellow G; Acid Yellow G pat.; Fast Yellow 053.

Scientific name.—Amidoazobenzene-disulphonate with some sodium amidoazobenzene-monosulphonate.

Discovered.—1878.

Shade.—Yellow. Offered by 5 out of 12 sources.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd., April 27, 1886, p. 643), where it is classified among the "nontoxic" colors.
- 3. Fraenkel (p. 575), where it is stated to be nonpoisonous.
- 4. Permitted by the law of Austria.

### UNFAVORABLE.

- 1. Weyl (p. 115): "Poisonous to human beings. (?)"
- 2. Chlopin (p. 151) considers that the work of others makes this a suspicious color. His own experimental data are as follows:

Experimental data by Chlopin.

G. T. 8 AND 9.

[1 gram=152 mg=106 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Mar. 11-13 14 15 16 17 18 19 20 21	3 4	6. 6 6. 6	310 310 305 380 320 280 292	Dog normal; color urine normal; acid; no albumen. Do. Do. Urine greenish brown; reddens with H <sub>2</sub> SO <sub>4</sub> and HCl; no albumen. Color same; traces of albumen. Color less intense; trace of albumen. Do. Urine greenish brown; albumen gone; dog is lively. Everything normal.

Conclusion: "Suspicious."

- 3. Buss (Forschungsber. über Lebensmittel, 1896, Vol. III, p. 173): Is regarded as poisonous.
- 4. Kobert (Lehrbuch der Intoxicationen, 1893, p. 335): Listed as poisonous.
- Lewin (Lehrbuch der Toxikologie, 1897, p. 231) says "produces eczema," and cites Deutsche Med. Wochenschr., 1891, p. 45.

## G. T. 9.

Trade names.—Fast Yellow R; Fast Yellow; Yellow W.

Name under which it was offered on the United States market as a food color in 1907.—Fast Yellow 034.

Scientific name.—Sodium salt of amidoazotoluene-disulphonic acid. Discovered and patented.—1878.

Shade.—Yellow. Offered by 1 out of 12 sources.

#### FAVORABLE

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 31): " \* \* \* not poisonous to human beings and dogs \* \* \* \* Solid Yellow."
- 3. Cazeneuve and Lépine (Compt. rend., 1885, v. 101, pp. 1167-1169): A. A dog received 42 milligrams per kilogram of body weight, or 29 grains per 100 pounds, for 5 days; thereupon received four times that amount for 5 days, or 168 milligrams per kilogram body weight, or 117 grains per 100 pounds; for the 10 days next succeeding it received daily twice the last dose, or 336 milligrams per kilogram of body weight, that is, 235 grains per 100 pounds; it then received in 1 day 20 times the original dose, or 840 milligrams per kilogram body weight, that is, 596 grains per 100 pounds, and during the entire period nothing abnormal was noticed. B. Three chronic invalids received from 2 to 4 grains of the dye daily; except colic without diarrhea nothing abnormal. They concluded that this dye is no more harmful than Naphthol Yellow S (G. T. 4).
- CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd., 1886, p. 643): Tolerated by man, well or sick.

### UNFAVORABLE.

- 1. Weyl (p. 115): "Poisonous to human beings. (?)"
- 2. Chlopin (p. 151): Where he considers that the work of others makes this a suspicious color. For his experimental data thereon see table under G. T. 8; Chlopin's chemical description of the dye used applies to both G. T. 8 and 9.
- 3. Kobert 'Lehrbuch der Intoxicationen, 1893, p. 336): Listed as poisonous.

## G. T. 11.

Trade names.—Sudan I; Carminaph.

Names under which it was offered on the United States market as a food color in 1907.—Oil Orange 7078; Cerasin Orange I.

Scientific name.—Benzene-azo-betanaphthol.

Discovered.—1883.

Shade.—Orange Yellow. Offered by 2 out of 12 sources.

### FAVORABLE.

- 1. Weyl (p. 115): "Nonpoisonous \* \* \* Soudan I \* \* \*."
- 2. "Other Azo-colors, \* \* \* for instance Soudan I \* \* \* are entirely non-poisonous." (p. 148.)

1. Prohibited by Confectioners' List.

- 2. Weyl (p. 119): Dog received 18 grams in 6 doses in 17 days, and remained under observation 5 days longer; 4 doses of 168 milligrams per kilogram body weight (118 grains per 100 pounds) and 2 doses of 420 milligrams per kilogram body weight (294 grains per 100 pounds) produced colored urine, phenol in urine, vomiting and distinct albuminuria beginning with the third dose. From the foregoing Weyl concludes as follows: "The color in the doses administered is not entirely harmless, since a limited albuminuria seems to be brought about."
- 3. Fraenkel (p. 576): "It is not wholly harmless, since this coloring matter seems to produce a slight albuminuria."

## G. T. 13.

Trade names.—Ponceau 4 G B; Crocein Orange; Brilliant Orange; Orange G R X.

Names under which it was offered on the United States market as a food color in 1907.—Crocein Örange 10234; Crocein Örange; Crocein Örange G; Ponceau 4 G B.

Scientific name.—Anilin-azo-betanaphthol-monosulphonic acid. Discovered.—1878.

Shade.—Orange Yellow. Offered by 6 out of 12 sources.

#### FAVORABLE.

1. Permitted by Confectioners' List.

2. Weyl (p. 115): "Nonpoisonous \* \* \* Ponceau 4 G B \* \* \*."

3. Experiment on dog in doses of 161 milligrams per kilogram body weight; that is, 113 grains per 100 pounds body weight, apparently produced no disturbance aside from coloring of the urine. (p. 124.)

4. Weyl (p. 148): "Other Azo-colors \* \* \* for instance \* \* \* New Coccin \* \* \* are entirely nonpoisonous."

5. Weyl's conclusion reads as follows: "This color can be regarded as nonpoisonous."

6. Fraenkel (p. 577): "Ponceau 4 G B can be regarded as nonpoisonous."

#### UNFAVORABLE.

1. Excluded by law of Austria.

## G. T. 14.

Trade names.—Orange G; Orange G G.

Names under which it was offered on the United States market as a food color in 1907.—Orange G G crystals; Orange G.

Scientific name.—Anilin-azo-betanaphthol-disulphonic acid G.

Discovered and patented.—1878.

Shade.—Orange Yellow. Offered by 2 out of 12 sources.

1. Chlopin examined this color, and his experimental data are as follows:

## Experimental data by Chlopin.

No. 1 (p. 123).

[1 gram=73 mg=51 grains.]

Date.	Dose.	Weight.	24-hours' urine.	General condition of animal and urine.
1901. Apr. 4 5 6 7 8 9 10	Grams. 1 2 2 3	Kilos. 13.7	cc. 525 600 990 550 680 730 755	Nothing abnormal; no albumen. Urine light chocolate brown, acid; no albumen. Urine clear, yellow, blackish sheen; no albumen. Urine clear, dark brown; no albumen; faintly alkaline. Do. Nothing abnormal. Do.

No. 2 (p. 124).

[1 gram=119 mg=103 grains.]

1901. Nov. 8	othing
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Conclusion: Nonpoisonous.

UNFAVORABLE.

1. Excluded by Austrian law.

## G. T. 15.

Trade names.—Ponceau 2 G; Orange R.

Scientific name.—Sodium salt of benzene-azo-beta-naphthol-disulphonic acid R.

Shade.—Bright Red. Not offered.

#### FAVORABLE.

1. Permitted by Confectioners' List.

#### UNFAVORABLE.

- 1. Excluded under Austrian law.
- 2. Excluded under Swiss laws.

## G. T. 16.

Trade names.—Butter Yellow; Oil Yellow.

Scientific name.—Dimethyl-amido-azo-benzene.

Discovered.—1875.

Shade.—Yellow. Not offered.

1. Weyl (p. 31): "Butter Yellow produces no disturbance in rabbits."

#### UNFAVORABLE.

1. Chlopin (p. 138): Where as a result of his own experiments he considers it poisonous. His experimental data are as follows:

Experimental data by Chlopin.

## No. 1.

### [1 gram=159 mg=111 grains.]

Date.	Dose.	Weight.	General condition of animal and urine.
1902. May 6 7-8 9-15 16 17 18	Grams.	Kilos. 6.3	Before experiment urine normal; after a few hours vomiting; urine not collected. No vomiting; eats poorly; more tired than usual. Dog gradually becomes normal. Vomiting; loss appetite. Repeated vomiting. Vomiting continues.

#### No. 2.

#### [1 gram=96 mg=67 grains.]

1903. Feb. 5 6	2 3	10.4	Before experiment dog quite normal; acid; no albumen.  Vomiting during night; dog does not eat, but drinks; urine more yellow than normal; acid; no albumen; in the evening the dog walked and drank water.  Died during the night; thin excreta in kennel; autopsy showed paralysis of heart as causing death.
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## No. 3.

	[1 gram=167 mg=117 grains.]					
1903.						
Feb. 8	2 2	6	Five hours after administration vomiting and involuntary thin feces.			
9			Dog does not eat; drinks much; weak; urine dark yellow; no vomiting.			
10			Dog lies in cage; moves slowly when allowed out of cage; in the evening			
11		•••••	retching.			
12			Dog is still weak, but general condition somewhat better; urine strikingly			
12			colored; acid; no albumen.			
13			Began to eat; no albumen.			
14-16			General condition improving and almost normal; lassitude continues			
			more than usual; urine normal color; acid; no albumen.			
17	3 2		In the evening repeated vomiting; loss of apetite.			
18	2		No vomiting; in the evening bothersome; does not eat.			
19		5.7	Stools normal; striking disturbance of movements; paralysis of legs; must			
			spread hind legs to stand; when pushed not only falls, but turns on his back; sight and hearing are normal; dog died at night.			

<sup>1</sup> This dog ran away.

## G. T. 17, 18, 41.

17. Trade names.—Chrysoidin Y; Chrysoidin crystals.

Name under which it was offered on the United States market as a food color in 1907.—Chrysoidin Y.

Scientific name.—Anilin-azo-meta-phenylene-diamin.

Discovered.—1875.

<sup>&</sup>lt;sup>2</sup> Gruebler's make.

<sup>&</sup>lt;sup>3</sup> Berlin make.

Shade.—Orange. Offered by 2 out of 12 sources.

18. Trade names.—Chrysoidin R; Cerotin Orange; C extra; Gold Orange for cotton.

Name under which it was offered on the United States market as a food color in 1907.—Chrysoidin R.

Scientific name.—Anilin-azo-meta-tolylene-diamin.

Shade.—Yellow brown. Offered by 1 out of 12 sources.

41. Trade name.—Chrysoidin R.

Scientific name.—Hydrochlorid of toluene-azo-meta-tolylene-diamin.

Discovered.—1876.

Shade.—Orange Brown.

(Note.—It has not been possible accurately to differentiate in the literature as to whether Nos. 17, 18, or 41, or all three, were referred to.)

#### FAVORABLE.

- 1. Weyl (p. 115): "Nonpoisonous \* \* \* Chrysoidin \* \* \*."
- 2. Permitted by the law of Italy.

#### UNFAVORABLE.

1. Prohibited by Confectioners' List.

2. Weyl (p. 126): Experiment on dog, giving him 113 milligrams per kilogram body weight, or 79 grains per 100 pounds body weight, produced a slight albuminuria.

In a second experiment a dog receiving "1 grain" ("grain" is evidently a misprint for "gram" and will be so treated), that is, 105 milligrams per kilogram body weight, or 74 grains per 100 pounds body weight, daily for one month, did not produce albuminuria, but caused a loss of body weight of about 12.5 per cent. A third dog receiving about 2 milligrams per kilogram body weight, or about 1.5 grains per 100 pounds body weight subcutaneously, suffered a loss of 20 per cent of its body weight in 23 days; dose repeated 24 days after and animal kept under observation seven days longer when animal was normal.

3. Weyl in summarizing experiments on these three dogs concludes as follows: "Chrysoidin produces, according to my investigations, a slight albuminuria, and notable reduction in body weight, but further disturbance has not been noted." (p. 127.)

4. Fraenkel (p. 577): "The above-mentioned Chrysoidin \* \* \* produces a slight albuminuria, and a notable decrease of body weight, and produces factory

eczema."

 Lewin (Lehrbuch der Toxikologie, 1897, p. 231): "Produces eczema," and cites Deutsche Med. Wochenschr., 1891, p. 45.

## G. T. 28.

Trade name.—Archil Substitute V.

Scientific name.—Sodium salt of para-nitrobenzene-azo-alpha-naph-thylamin-para-sulphonic acid.

Discovered and patented.—1878.

Shade.—Red. Not offered.

1. Permitted by Confectioners' List.

2. Weyl (p. 115): "Nonpoisonous \* \* \* Archil Substitute \* \* \*."

Describing experiments on three dogs, as follows: A. 430 milligrams per kilogram body weight, or 301 grains per 100 pounds body weight, on each of two successive days, and double the dose on the fourth day, producing no vomiting, but a tendency to vomit, a slight albuminuria and colored urine. B. 182 milligrams per kilogram body weight, or 127 grains per 100 pounds body weight, administered daily for one month; results similar to foregoing, but no colored urine. C. 105 milligrams per kilogram body weight, or 116 grains per 100 pounds body weight administered subcutaneously, produced only slight albuminuria, and no reduction in body weight. (p. 125.)

## G. T. 43.

Trade names.—Orange GT; Orange RN; Orange O; Orange N. Scientific name.—Sodium salt of toluene-azo-beta-naphthol-sulphonic acid.

Discovered.—1879.

Shade.—Orange. Not offered.

#### FAVORABLE.

1. Permitted by Confectioners' List.

#### UNFAVORABLE.

- 1. Excluded by Austrian law.
- 2. Excluded by Swiss laws.

## G. T. 55.

Trade names.—Ponceau R; Ponceau 2 R; Ponceau G and GR; Xylidin Red; Xylidin Scarlet.

Names under which it was offered on the United States market as a food color in 1907.—Scarlet; Orange R.

Scientific name.—Sodium salt of xylene-azo-beta-naphthol-disulphonic acid.

Shade.—Scarlet. Offered by 2 out of 12 sources.

### FAVORABLE.

1. Permitted by Confectioners' List.

2. Weyl (p. 31): "According to Cazeneuve and Lépine's experiments the following are not poisonous to human beings and dogs \* \* \* Ponceau R \* \* \*."

"Ponceau R (Ponceau 2 R, Xylidin Red, Xylidin Ponceau), not poisonous to dogs
neither by administration by stomach nor injection into blood." (p. 115.)

4. "Other Azo colors \* \* \* Xylidin Red \* \* \* are entirely nonpoisonous."
(p. 148.)

5. Fraenkel (p. 575): "That the monazo coloring matters examined by Cazeneuve and Lépine, as already above stated, were nonpoisonous, can be easily explained by the constitution of these substances. These two investigators examined \* \* \* Ponceau R \* \* \*."

- 6. Lieber (p. 140): A guinea pig received 310 milligrams per kilogram body weight or 217 grains per 100 pounds body weight, once a day six times every other day; the appeared to remain good, and no disturbances were noted.
- 7. Permitted by Austrian law.
- 8. Permitted by Swisslaws.
- 9. CAZENEUVE and LÉPINE (Bull. de l'acad. de méd., 1886, p. 643): Tolerated by man well or sick.
- 10. Chlopin (p. 150) classes it as nonpoisonous on his own experiments. His experimental data are as follows:

## Experimental data by Chlopin.

### [1 gram=141 mg=99 grains.]

Date.	Dose.	Weight.	24-hours' urine.	General condition of animal and urine.
1902. May 3 4 5 6 7 8 9	Grams. 2 2 2	Kilos. 7.1	cc. 370 351 420 290 359 360 330	Dog and urine normal and no albumen. Urine rose-colored; no albumen. Color normal; no albumen. Rose-colored urine; no albumen. Normal color; no albumen. Rose-colored; no albumen. Color and composition normal.
Total.	6			

#### · UNFAVORABLE.

- Prohibited by the ordinance of the police commissioner in France. (See Lieber, p. 30.)
- 2. MEYER (J. Amer. Chem. Soc. 1907, v. 29, pp. 900-901): The dog experimented on showed signs of paralysis on the morning of the seventh day at 8 o'clock, and died at 10.40 a. m., after having received a total of 32 grams of color, of which 16 had been given on the last day. The initial dose was 70 grains per 100 pounds body weight; the total weight of color was 5,818 milligrams per kilogram body weight, or 4,073 grains per 100 pounds; the average daily dose was therefore 582 grains per 100 pounds, or 831 milligrams per kilogram body weight.

#### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 65.

Trade names.—Fast Red B; Bordeaux B; Bordeaux BL; Bordeaux R extra.

Names under which it was offered on the United States market as a food color in 1907.—Bordeaux B; Claret Red.

Scientific name.—Alphanaphthylamin-azo-betanaphthol-disulphonic acid.

Discovered.—1878.

Shade.—Red. Offered by 2 out of 12 sources.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 115): "Bordeaux Red (Fast Red B) not poisonous to human beings."
- 3. "Other Azo colors \* \* \* Fast Red B \* \* \* are entirely nonpoisonous." (p. 148.)
- 4. Fraenkel (p. 575): "That the monoaco coloring matters examined by Cazeneuve and Lépine, as already above stated, are nonpoisonous, can be easily explained by the constitution of these substances. These two investigators examined \* \* \* Bordeaux B."
- 5. Arloing and Cazeneuve (Archives de physiologie, 1887, pp. 356-393): As a result of this work, which is divided into three parts—(1) Stating the effect of direct introduction of the color into the circulation; (2) intravenous injections; (3) comparing the effects of injections of color and of salt; and (4) feeding by the mouth—these investigators conclude that these coloring matters are toxic only in extremely large doses; that when given to dogs with their food that no inconvenience of any kind results; this is based upon experiments on three dogs, covering 145 days, where each dog received per kilogram of initial body weight in the first case 20,307 milligrams, or 14,213 grains per 100 pounds initial body weight; in the second case, 29,590 milligrams, or 20,713 grains per 100 pounds initial body weight. Per day this means 98 grains per 100 pounds initial body weight in the first case; in the second case, 143 grains per 100 pounds initial body weight; and in the third case, 137 grains per 100 pounds initial body weight.
- 6. CAZENEUVE (Arch. gén. de méd., 1886, p. 753) says it may be taken without effect by man or animals, sick or well, in large doses.
- 7. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd., 1886, p. 643): Tolerated by man well or sick.

## G. T. 70.

Trade name.—Azarin S.

Scientific name.—Ammonium bisulphite compound of dichlorophenol-azo-beta-naphthol.

Shade.—Red. Not offered.

#### FAVORABLE.

1. Permitted by Confectioners' List.

2. Weyl (p. 115): "Nonpoisonous \* \* \* Azarin S \* \* \*."

- 3. Experiments on five dogs; three fed by the mouth; two treated hypodermically, of which latter one died. The first dog received 1,367 milligrams per kilogram body weight in 25 days; that is, 54.7 kilograms per day on the average, or a total of 957 grains per 100 pounds of body weight; that is, 38 grains per 100 pounds body weight per day. The second dog received a total of 1,942 milligrams per kilogram body weight in 20 days, or 97 milligrams per kilogram body weight per day, which amounts to a total of 1,359 grains per 100 pounds body weight, or 68 grains per 100 pounds body weight per day. In both cases a distinct amount of albumen was present in the urine, and the urine evolved sulphurous acid on treatment with hydrochloric acid. The third dog received hypodermically three doses in eight days, each dose being 213 milligrams per kilogram body weight; that is, 149 grains per 100 pounds body weight; no bad effects. (p. 133.)
- 3. "Administered by the stomach Azarin S is harmless." (p. 134.)
- 4. "Other Azo colors, \* \* \* for instance Azarin S, are entirely nonpoisonous."
  (p. 148.)
- 5. Fraenkel (p. 578): "Azarin S administered by the stomach is entirely harmless."

- 1. Weyl (p. 134): Dog; weight not given; received 5 cc of Azarin S paste by injection into the abdominal cavity, and survived three days. "The cause of death was considered to be peritonitis without effusion. The result of this postmortem is of much interest. The red spots consisted, as was determined by chemical analysis, of the azo color which is the basis of the Azarin S. Consequently in the peritoneal cavity the same splitting up of the Azarin S had occurred which takes place when it is attached to textiles."
- 2. Fraenkel (p. 578): To the same effect.

## G. T. 78.

Trade name.—Erika B.

Scientific name.—Sodium salt of methyl-benzenyl-amido-thio-xylenol-azo-alpha-naphthol-disulphonic acid.

Discovered and patented.—1889.

Shade.—Rose Pink. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 153): Based on his own experiments considers it as not harmless. The experimental data are as follows:

### Experimental data by Chlopin.

[1 gram=125 mg=87.5 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 9 10 11 12 13 14 15 16 17 18	2 2 6	Kilos. 8.0	cc. 392 400 420 360 390 293 350 390 402 350	Dog normal; urine acid; no albumen. Urine of rose shade; insignificant traces of albumen; acid. Color same; no albumen. Do. Urine yellow, greenish shade; traces of albumen. Color normal; no albumen. Do. Urine wine yellow, orange, acid; no albumen. Dog quite well; urine orange; no albumen.

## G. T. 84.

Trade names.—Resorcin Yellow; Tropæolin O; Tropæolin R; Chrysoin; Chryseolin; Yellow T; Gold Yellow; Acme Yellow.

Names under which it was offered on the United States market as a food color in 1907.—Chrysoin REZ; Resorcin 0275.

Scientific name.—Sodium salt of para-sulphobenzene-azo-resorcinol. Discovered.—1875.

Shade.—Reddish yellow. Offered by 2 out of 12 sources.

1. Chlopin (pp. 131-2) examined this color physiologically, and has classified it as harmless. The experimental data are as follows:

Experimental data by Chlopin.

No. 1 (p. 227).

[1 gram=43 mg=30 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 9	Grams.	Kilos. 23.00	cc. 500	Dog quite well; urine normal color, acid; no albumen.  Do. Urine acid; no albumen.
11 12 13 14 17	1 2.00		460 520 580	Ornic acid, no aboutmen.  No symptoms of poisoning; eats.  Urine dark brown; no albumen.  Do.  Urine normal color; no albumen; dog is well.
Total	1 3. 21 2 0. 70	·		

No. 2.

### [1 gram=156 mg=109 grains.]

1901. Oct. 9 10 11 12 13 14 15 16 17 18 19 20 21 22-26 Total	2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2 2	7.0	300 350 335 290 290 375 300 360 360	Dog well; urine yellow; no albumen. Do. Urine brown, acid; no albumen. Urine light brown, acid; no albumen. Do. Do. Do. Do. Do. Urine light brown, acid, no albumen; diarrhea. Brown yellow, acid, no albumen no diarrhea. Urine normal; dog is well. Do.
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<sup>1</sup> Internally.

<sup>2</sup> Subcutaneously.

2. Permitted by the law of Italy.

#### UNFAVORABLE.

1. Forbidden by the Confectioners' List.

#### DOUBTFUL.

1. WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; not indifferent.

## G. T. 85.

Trade names.—Orange I; Alphanaphthol Orange; Naphthol Orange; Tropæolin OOO; Orange B.

Names under which it was offered on the United States market as a food color in 1907.—Orange RZ; Orange 027.

Scientific name.—Sodium salt of para-sulphobenzene-azo-alphanaphthol.

Discovered.—1876.

Shade.—Orange. Offered by 2 out of 12 sources.

1. Permitted by Confectioners' List.

2. Weyl (p. 31): "According to Cazeneuve and Lépine's experiments, the following are not poisonous to human beings and dogs: \* \* \* Orange \* \* \*."

3. "Orange I (Alpha-naphthol Orange, Tropæolin OOO) not poisonous to dogs neither by administration by stomach, nor by injection into blood." (On authority of Cazeneuve and Lépine.) (p. 115.)

4. Weyl (pp. 123, 148) refers to this as not poisonous.

5. Permitted by the law of Italy.

6. Permitted by the law of Austria.

7. CAZENEUVE (Arch. gén. de méd., 1886, Vol. I, p. 753) says it may be taken without effect by man or animals, sick or well, in large doses.

8. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd., 1886, p. 643): Tolerated by man, well or sick.

## G. T. 86.

Trade names.—Orange II; Betanaphthol Orange; Tropæolin OOO No. 2; Mandarin G extra; Chrysaurein; Gold Orange; Orange extra; Atlas Orange; Orange A.

Names under which it was offered on the United States market as a food color in 1907.—Naphthol Yellow SLOZ; Orange II; Orange; Orange Y; Mandarin G extra; Orange A 1201; Orange A extra.

Scientific name.—Sodiúm salt of para-sulphobenzene-azo-beta-naphthol.

Discovered.—1876.

Shade.—Orange. Offered by 8 out of 12 sources.

#### FAVORABLE.

1. Permitted by law in Italy.

2. Frentzel (Zts. Nahr. Genussm., 1901, v. 4, p. 974) says that according to his experiments this color, in the small amounts in which it is used in food products and which can enter the human system in the course of 24 hours, can hardly, even with frequent administration, cause a harmful effect.

3. Frentzel (Zts. Nahr. Genussm., 1901, v. 4, pp. 968-974): Experimented on rabbits, giving dye with food, a total of 21 grams in 19 days, a total of 8,748 milligrams per kilogram body weight, or 6,133 grains per 100 pounds body weight, in doses of 1 gram each, daily for the first 15 days, or about 417 milligrams per kilogram of body weight, or 292 grains per 100 pounds body weight per dose for these 15 doses; the color could only be detected in the urine, and the feces became softer. A dog was given 1,020 milligrams per kilogram body weight, or 714 grains per 100 pounds body weight, and showed distinct kidney irritation, great thirst, and diarrhea; recovery required about one week; and thereafter the same animal was fed by the mouth one-twentieth of the above dose each day for 9 successive days without any untoward effect. A second dog received per kilogram of body weight 172 milligrams, or 121 grains per 100 pounds body weight, and it, like the first dog, in the first experiment, showed kidney irritation, diarrhea, and great thirst. On humans 100 milligrams, or 11 grains, colored the urine within 15 minutes, and this color remained for 24 hours; there was no sign of vomiting or diarrhea; the bitter taste of the color was noticeable.

1. Forbidden by Confectioners' List.

2. Forbidden by Swiss Analytical chemists.

3. Weyl (p. 115): "Poisonous Orange II. \* \* \*."

4. "Betanaphthol Orange is, therefore, according to Experiment I, poisonous in small doses when administered by the stomach, and suffices to kill an ordinarily

large strong dog." (p. 123)

5. A. Weyl's own experiments on two dogs, the initial dose in one case being 476 milligrams per kilogram body weight, or 333 grains per 100 pounds body weight; the animal receiving in the course of 20 days, in four doses, 1,333 milligrams per kilogram body weight, or 933 grains per 100 pounds body weight; or 335 milligrams per dose per kilogram body weight; that is, 225 grains per dose per 100 pounds body weight. The animal died, and suffered diarrhea and albuminuria, and its urine was colored red throughout the entire period. B. The second dog received hypodermically per kilogram body weight, 116 milligrams, or 61 grains per 100 pounds body weight; its urine was colored; albuminuria, diarrhea, loss of hair, abscesses, and loss of weight occurred. It required 36 days to recover from four doses administered during one week. C. A rabbit received 1,333 milligrams per kilogram body weight, or 933 grains per 100 pounds body weight, and died within 12 hours. (p. 122)

6. "Of the 23 Azo colors subjected to examination only two \* \* \* \* Orange II produce(s) such effects when administered by the stomach that we can consider it poisonous. With dogs the lethal dose is less than 1 gram per kilo of

the body weight of Orange II \* \* \*." (p. 147.)

7. "Further Orange II, which is poisonous \* \* \*."

8. "The poisonous qualities of Orange II."

9. "Further, in spite of the presence of the sulpho groups, colors may be poisonous, as is shown with Orange II." (p. 148.)

10. Chlopin (Zts. Nahr. Genussm., 1902, v. 5, p. 241): A. A dog received 349 milligrams per kilogram body weight, or 244 grains per 100 pounds body weight for the first day of experiment, which dose was repeated on the third and fourth days; nothing untoward is noted for the first three days in the condition of the dog: the urine was dark red but free from albumen; on the fifth and sixth days two-thirds of the above amount was given, and on the seventh and eighth days the original dose was given. On the fourth day the animal was frisky and had a good appetite but was vomiting; on the fifth day vomiting stopped, but diarrhea ensued, which diarrhea continued for one week; the weight remained practically constant; the urine was colored throughout from dark red to orange red and dark brown and became normal the fourth day after the last administration. B. Humans: Chlopin took 200 milligrams, or  $3\frac{1}{12}$  grains, in a gelatin capsule at 3 p. m.; at 4.30 p. m. the urine was colored a strong red orange; at 6 p. m. a dryness of throat and bad taste in mouth appeared; at 6.30 p. m. felt very badly; vertigo and unable to remain seated and continue writing; blood rushed to head; the general condition very poor; somewhat improved by moving about in open air; 7.30 felt so poorly took Glauber's salt as an antidote; 11 p.m. ill condition still continuing; urine normal yellow; midnight recovered.

Chlopin states that he would not repeat this experiment on himself, or on any other human, and he concludes therefore that this color must be regarded as harmful.

11. Chlopin (p. 133) classes it as "harmful." The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram=116 mg=81 grains.]

Date.	Dose.	Weight.	24-hours' urine.	General condition of animal and urine.				
1901. May 27 28 29 30 June 31 June 1 2 3 4 5 6 8	Grams. 3 3 3 2 2 2 3 3 3	**************************************	cc. 350 340 350 350 350 325 330 350 315 375	Dog is quite well; urine normal. Urine brown red, acid; no albumen. Do. Diarrhea and vomiting at night, no albumen; urine orange red, acid, no albumen. Diarrhea continues; no albumen; no vomiting. Diarrhea continues; dog lively, eats with relish; no albumen. Urine brown orange, acid, no albumen; diarrhea continues. Urine clear; no albumen; diarrhea continues. Urine dark brown; no albumen; diarrhea decreases. Diarrhea still less; no albumen. Diarrhea very slight; urine slightly orange, no albumen. Urine normal in color and composition; diarrhea stopped.				
Total	19							

12. Buss lists it as poisonous.

DOUBTFUL.

1. WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

G. T. 87.

Trade names.—Orange III; Helianthin; Tropæolin D; Methyl Orange; Dimethylanilin Orange.

Scientific name.—Sodium salt of para-sulphobenzene-azo-dimethy-lanilin.

Discovered.—1876.

Shade.—Orange Yellow. Not offered.

FAVORABLE.

1. Permitted by the law of Italy.

UNFAVORABLE.

1. Chlopin (pp. 145, 146) on his own experiments classes it as poisonous. The experimental data are as follows:

Experimental data by Chlopin. No. 1.

[1 gram=163 mg=114 grains.]

Date.	Dose.	Weight.	24-hours' urine.	General condition of animal and urine.
1901. Aug. 29 30 31 Sept. 1 2 3 4 5 6 7 8-9	2 2 2 2 2 2	Kilos. 6.14	cc. 310 250 330 305 305 Little. Little. Little.	Before experiment dog quite normal; color urine usual, acid, no albumen. Do. Color of urine dark yellow; no albumen. Urine brown; reddish with sulphuric acid; no albumen. Do. Do. Do. Do. Urine dark brown; no albumen; vomiting. Paresis of hind legs; dog does not eat, but drinks with avidity. Complete ataxia, which became general on the 9th; dog can not move in straight line, and walks in circles, and drops his head on things and falls; after falling arises with difficulty and stands with widely spread feet, continuously lifting now one then the other; extremities shake; on 11th day sat with difficulty; laced on his feet, maintained equilibrium with difficulty; involuntarily lifts one paw and moves it about in the air; animal can still swallow; eyes respond to light; does not eat; on the 12th the hind legs completely paralyzed; on the 14th vomiting and paralysis of the front legs; lies quietly without moving; is killed. Autopsy shows no change in internal organs except hyperæmia in the lumbar region of the "spinal column (?) bordering between anterior and lateral columns," causing death by paralysis of the heart.
Total	10			comme, county source of paralysis of the heart.

## Experimental data by Chlopin—Continued.

#### No. 2

#### [1 gram=149 mg=104.3 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Dec. 3 4 5	Grams. 3	Kilos. 6.7	cc. Little. Little.	A few hours after giving color dog in tremors; does not eat. Urine black; acid; no albumen; paralysis of the extremities. Dog lies in cage in full paralysis; died before dinner; no albumen in urine; cause of death, paralysis of heart.

2. Meyer (J. Amer. Chem. Soc., 1907, v. 29, p. 900): Dog receiving 113 milligrams per kilogram body weight; that is, 79 grains per 100 pounds body weight. Result, diarrhea, which continued throughout 17 days, although only 3 doses of the same size were given in 6 days; thereafter the dose was increased geometrically; the diarrhea continued; the coloring matter was discharged in the urine and the feces; there was no abnormal condition revealed by the autopsy.

#### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 88.

Trade names.—Diphenylamin Orange; Orange IV; Tropæolin OO; Orange M; Fast Yellow; Orange G S; New Yellow; Orange N; Acid Yellow D.

Scientific name.—Sodium salt of para-sulphobenzene-azo-diphenylamin.

Discovered.—1876.

Shade.—Orange Yellow. Not offered.

### FAVORABLE.

- 1. Weyl (p. 115): "Nonpoisonous \* \* \* Diphenylamin Orange \* \* \*
- 2. \* \* \* Diphenylamin Orange is \* \* \* nonpoisonous." (p. 132.)
- 3. "For instance, the poisonous Metanil Yellow corresponds to the nonpoisonous Diphenylamin Orange." (p. 148.)
- 4. The experiments on which Weyl based the conclusions above may be summarized as follows: A. A dog received 183 milligrams per kilogram body weight, or 128 grains per 100 pounds body weight. The urine was rendered black, and contained traces of phenol and abundant albumen; this condition lasted for 72 hours, at the end of which 110 milligrams per kilogram body weight, or 77 grains per 100 pounds body weight, were administered; urine became colorless and albumen diminished. Five days afterwards a total of 10 grams, or 366 milligrams per kilogram body weight, or 256 grains per 100 pounds body weight, were administered; the urine became abundant, was strongly black, alkaline, contained albumen. The loss of weight was 1 kilogram, or about 3½ per cent. B. A dog received 308 milligrams per kilogram, or 216 grains per 100 pounds body weight, as the initial dose; albuminuria did not result until after repeated dosing same as initial dose, and administered for 2 weeks.
- 5. Weyl's conclusions are as follows: "According to the above investigations, Diphenylamin Orange causes albuminuria, but further disturbances did not appear during the several weeks' observations on the animals used."

6. Chlopin (p. 148) examined this color and classes it as nonpoisonous. The experimental data are as follows:

### Experimental data by Chlopin.

#### [1 gram=145 mg=100 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Oct. 27	Grams.	Kilos.	cc. 300	Dog quite normal; urine color normal; no albumen. Urine dark brown; acid; no albumen.
29 30 31 Nov. 1	3 3 3 3			Do. Do. Do. Urine yellow, with sulphuric acid red; no albumen.
2 3 4	3 3	6.5	340 275	Orange with orange sheen; no albumen. Do. Urine brown yellow; acid; no albumen.
5 6	3	6.7	300	Do. Do.
Total.	24			

- 7. Fraenkel (pp. 577, 578) to the same effect as Weyl, as above.
- 8. Permitted by the law of Italy.

#### UNFAVORABLE.

1. Prohibited by Confectioners' List.

## G. T. 89.

Trade names.—Brilliant Yellow S; Yellow WR; Curcumin.

Names under which it was offered on the United States market as a food color in 1907.—Brilliant Yellow S.

Scientific name.—Sodium salt of para-sulphobenzene-azo-diphenyl-amin-suphonic acid.

Shade.—Yellow. Offered by 1 out of 12 sources.

#### FAVORABLE.

Lieber (p. 136): A very young rabbit received six doses on alternate days, each
dose amounting to 320 milligrams per kilogram of body weight, or 224 grains per
100 pounds of body weight. No untoward symptoms are recorded; the body
weight increased almost 10 per cent in 11 days.

UNFAVORABLE.

Nothing.

## G. T. 92.

Trade names.—Azo Acid Yellow; Azoflavin; Azo Yellow; Indian Yellow.

Scientific name.—Mixture of nitrated diphenylamin yellow with nitro-diphenylamin.

Discovered.—1880.

Shade.—Yellow. Not offered.

97291°-Bull, 147-12-7

1. Chlopin (p. 128) examined this color and classifies it as nonpoisonous. The experimental data are as follows:

Experimental data by Chlopin.

No. 1.

[1 gram=125 mg=87.5 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 6 8 10 11 12	Grams. 2	Kilos. 8	cc. 370 Little. 600 430	Dog normal; urine acid; no albumen. Urine reddish orange; acid; no albumen. Do. Urine red; no albumen. Urine almost normal color; acid; no albumen.
Total .	5			

Conclusion: Nonpoisonous.

No. 2 (p. 129).

#### [1 gram=143 mg=100 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 12 14 15	Grams.	Kilos.	cc. 430 610 600	Dog normal; urine acid; no albumen. Do. Using dark vallow; said; no albumen
Total.	5		400	Urine dark yellow; acid; no albumen. Urine almost normal color; acid; no albumen.

Conclusion: Showed no harmful effects.

2. Permitted by law in Italy.

UNFAVORABLE.

Nothing.

DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion; noticeably retards digestive action; is not indifferent.

## G. T. 93.

Trade name.—Azo-fuchsin G.

Scientific name.—Sodium salt of para-sulphobenzene-azo-dioxy-naphthalene-sulphonic acid.

Discovered.—1889.

Shade.—Reddish brown. Not offered.

FAVORABLE.

1. Chlopin (pp. 126, 127) examined this color and classifies it as nonpoisonous. The experimental data are as follows:

### Experimental data by Chlopin.

No. 1.

## [1 gram=117 mg= 82 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901 May 3	Grams.	Kilos. 8.5	cc. 300 363	Dog well and normal; urine acid; no albumen. Urine chocolate brown; acid; no albumen.
5 6	2		390 380	Do. Urine dark brown, chocolate, in thin layer greenish sheen; acid; no albumen.
7 8 9	2	8.7	285 420 292	Urine slightly greenish; acid; no albumen.   Dark chocolate brown; acid; no albumen.   Urine normal; dog is well.
Total.	6			

#### No. 2.

### [1 gram=125 mg=87.5 grains.]

1901. Oct. 9 10 11 12 13 14 15 16 17 18 19 20 21	2 2 2 2 2 2	8.0	390 310 380 325 396 397 325 380 246 420	Dog quite normal; urine acid; no albumen.  Do. Urine slightly more yellow; no albumen. Urine dark brown; acid; no albumen.  Do. Do. Do. Do. Do. Urine almost black; acid; no albumen. Urine almost black; acid; no albumen. Urine almost black; acid; no albumen; urine of lighter color. Do.	
	12	8.4		Do.	
	1				

#### UNFAVORABLE.

Nothing.

#### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 94.

Trade names.—Tartrazin; Hydrazin Yellow.

Names under which it was offered on the United States market as a food color in 1907.—Flavazein Red Shade Z; Tartrazin; Acid Yellow AT.

Scientific name.—Sodium salt of benzene-azo-pyrazalone-carboxy-disulphonic acid.

Discovered.—1884.

Shade.—Yellow. Offered by 6 out of 12 sources.

### DOUBTFUL.

1. LIEBER (p. 134): Dog, 2 months old, received, per kilo, body weight, 37 milligrams, or 26 grains per 100 pounds body weight, six times, on alternate days. The animal suffered from mild diarrhea at the start, which continued with greater or less activity than at the start throughout the test.

- 2. Meyer (J. Amer. Chem. Soc., 1907, v. 29, p. 897): Dog received 100 milligrams per kilogram body weight, or 70 grains per 100 pounds, increased in geometric proportion for 6 consecutive days, at the end of which time diarrhea set in. On that day 2,000 milligrams per kilogram body weight, or 1,400 grains per 100 pounds body weight, were administered; this was about two-thirds as much as the animal had received in all the 5 days preceding; the feces were colored after the first administration, and the urine was also colored; albuminuria doubtful.
- 3. Fraenkel (pp. 210 and 216): Tartrazin, according to the Green Tables, is a derivative of isopyrazolon; "\* \* \* only those substances which are derivatives of pyrazolon are antipyretics, the isopyrazolon derivatives are, however, poisonous." Tartrazin is also closely related to the antipyrin class of compounds, which class is known to possess an irritant action and also a depressing action on the circulation. Tartrazin also contains benzol groups, which are said to increase the physiological activity of pyrazolon derivatives.

## G. T. 95.

Trade names.—Metanil Yellow; Orange MN; Tropæolin G.

Names under which it was offered on the United States market as a food color in 1907.—Victoria Yellow conc. Z; Yellow MXX conc.

Scientific name.—Sodium salt of meta-sulphobenzene-azo diphenylamin.

Discovered.—1879.

Shade.—Orange. Offered by 2 out of 12 sources.

#### FAVORABLE.

- 1. Frentzel (Zts. Nahr. Genussm., 1901, v. 4, p. 974): A. A rabbit received 379 milligrams per kilogram body weight, or 265 grains per 100 pounds body weight; no color administered for 2 days, and then administered at intermittent periods, so as to receive 6 doses in 19 days. Total weight administered per kilogram body weight 2,085 milligrams, or 1,460 grains per 100 pounds, which is equivalent to 77 grains per day per 100 pounds; the color could always be detected in the urine, and the feces became softer. B. A dog received 581 milligrams per kilogram body weight at one dose; that is, 407 grains per 100 pounds body weight. There was no vomiting and no diarrhea; the dye persisted in the urine for 72 hours. C. The same dog, after recovering from the foregoing, received daily one-tenth the above dose for 9 days; the dyestuff was found in the urine and the feces and movements were normal; dissection showed nothing abnormal. D. A human swallowed 100 milligrams, or 1.5 grains; no untoward symptoms are recorded; the urine remained colored for 24 hours. From the foregoing Frentzel concludes that this coloring matter is absolutely harmless.
- 2. Chlofin (Zts. Nahr. Genussm., 1902, v. 5, p. 241): A. A dog received 305 milligrams per kilogram body weight; that is, 104 grains per 100 pounds body weight; no color administered for four days; the same dose was given on alternate days six times, and the dose was increased 50 per cent, and that dose administered twice, and the original dose was given on the next alternate day; altogether the animal received per kilogram body weight, 3,355 milligrams, or 1,144 grains per 100 pounds body weight, in a period of 22 days, or an average of 153 milligrams per kilogram body weight per day, or 52 grains per day per 100 pounds body weight. At the end of the twelfth day albuminuria set in, and it required three weeks after ceasing the administration of the color for the albuminuria

2. Chlopin (Zts. Nahr. Genussm., 1902, v. 5, p. 241)—Continued.

to disappear; the urine was colored throughout the period of dosing, and it was not until 10 days after the last color had been administered that the urine regained normal color; otherwise the animal was well throughout. B. Humans: Two hundred milligrams, or  $3\frac{1}{12}$  grains taken at 3 p. m., October 30, 1901, colored the urine so highly yellow that it could be dyed with. Apart from the bitter taste of the product no untoward symptoms are recorded. C. Chlopin's conclusion ( $Zts.\ Nahr.\ Genussm.,\ 1902,\ v.\ 5,\ p.\ 244$ ): "Metanil Yellow is not poisonous to dogs in doses of from 2 to 3 grams per day, nor to humans in doses of 0.2 grams per day, and may therefore perhaps be regarded as non-poisonous from a practical point of view."

#### UNFAVORABLE.

- 1. Weyl (p. 115): "Poisonous \* \* \* Metanil Yellow."
- 2. Says the product smelled strongly of diphenylamin. (p. 130.)
- 3. A. "Metanil Yellow must be considered poisonous when administered by the stomach from the indications of Experiments 1 and 2. The lethal dose, which is determined by Experiment 2, is 0.53 grams per kilo body weight." This lethal dose is 371 grains per 100 pounds body weight. B. A dog received 862 milligrams per kilogram body weight, or 603 grains per 100 pounds body weight. This caused vomiting; the same dose was repeated 24 hours afterwards, the animal again vomiting. The animal died within 96 hours from the first administration. C. A dog received 89 milligrams per kilogram body weight, or 62 grains per 100 pounds body weight; the urine became colored about 96 hours after administration; 5 days after the first administration the animal was given 10 times the original dose; vomiting set in within one hour; in 24 hours the urine was deeper colored; a week later after the dose last preceding, one-half of that dose was given, and the animal died within 24 hours; the animal had lost during this period approximately one-quarter its original weight. (p. 132.)
- 4. "\* \* Metanil Yellow \* \* \* produce[s] such effects when administered by the stomach that we can consider them [it] poisonous." (p. 147.)
- 5. "The poisonous qualities of \* \* \* Metanil Yellow; the poisonous Metanil Yellow." (p. 148.)
- 6. Fraenkel (p. 578): "A dog weighing 11 kilograms was killed by 20 grams of this coloring matter within four days, whereas the isomeric Diphenylamin Orange is nonpoisonous, and it must first be considered if the poisonous nature of this substance can be explained by the easy liberation of diphenylamin from it, since this coloring matter, in and of itself, has a strong odor of diphenylamin."
- 7. CHLOPIN (p. 141): Based on his own experiments considers it "Not quite harmless." The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram = 153 mg = 107 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 18 19 20 21 22 23 24 25 26 27	2 2 2 2	Kilos. 6.55	cc. 390 350 308 300 370 380 322 320 365	Quite normal. Urine unusual color; acid; no albumen. Do. Urine orange; acid; no albumen. Do. Do. Do. Urine dark brown, no albumen. Do. Do.

## Experimental data by Chlopin—Continued.

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 28 29 30 31 June 1 2 3 4 5 6 7 8 9–29	Grams. 2 2 2 3 3 3 2 2	Kilos.	cc. 330 350 350 355 400 340 330 400 305 390 330 335	Urine dark brown, no albumen.  Do. Do. Do. Urine brown; traces of albumen. Do. Do. Do. Do. Do. Do. Do. Do. Do. Color of urine normal June 18, on 29th albumen disappears.
Total	22			

- 8. Chlopin (p. 141): On authority of others not stated, classes this color as harmful or poisonous.
- 9. Prohibited by the laws of Italy.
- 10. Prohibited by Confectioners' List.
- Prohibited by the Resolutions of the Society of Swiss Analytical Chemists, September, 1891.
- 12. Buss lists it as poisonous.

#### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says that it noticeably retards digestion.

## G. T. 97.

Trade names.—Orange T; Mandarin G R; Orange R; Kermesin Orange.

Name under which it was offered on the United States market as a food color in 1907.—Orange 2 R.

Scientific name.—Sodium salt of sulpho-ortho-toluene-azo-beta-naphthol.

Shade.—Orange. Offered by 1 out of 12 sources.

FAVORABLE.

Nothing.

### UNFAVORABLE.

Chlopin (p. 125): Based on his own experiments considers this color as "harmful." The experimental data are as follows:

### No. 1.

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 4 5 6 7 8 9	Grams.	Kilos.	cc. 310 280 420 570 370 250	Nothing abnormal. Urine reddish brown; traces of albumen; acid. Do. Urine dark brown; acid; traces of albumen. Color of urine weaker; no albumen. Almost normal-colored urine; no albumen; dog is well and lively. Nothing abnormal.
Total	4			

# No. 2. $[1 \ \mathrm{gram} = 217 \ \mathrm{mg} = 152 \ \mathrm{grains.}]$

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Jan. 25	Grams.	Kilos. 4.6	cc. 200	Soon after giving color heavy vomiting and diarrhea; dog does not eat or drink the rest of the day; urine acid; no albumen. Vomiting and diarrhea continued; dog is tired and run down, and began to eat; urine dark brown and no albumen.
27 28	2	4.3	180	Urine same; dog livelier. Diarrhea and vomiting stopped; urine of brown color; acid; no
29	2		200	albumen. Urine orange; no albumen; acid.
Total	6		. 8	

## G. T. 102.

Trade names.—Fast Red; Roccellin; Cerasin; Rubidin; Fast Red A; Rauracienne; Orcellin No. 4.

Scientific name.—Sodium salt of para-sulphonaphthalene-azo-beta-naphthol.

Shade.—Brownish red. Not offered.

Discovered and patented.—1877.

#### FAVORABLE.

- 1. Permitted by the law of Italy.
- 2. Permitted by the law of Austria.
- 3. CAZENEUVE (Arch. gen. de méd., 1886, p. 753) says it may be taken without effect by man or animals, sick or well, in large doses.
- 4. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd. 1886, p. 643): Tolerated by man well or sick.

## G. T. 103.

Trade names.—Azorubin S; Carmoisin; Azo Acid Rubin; Fast Red C; Azorubin A.

Names under which it was offered on the United States market as a food color in 1907.—Claret Red RZ; Cardinal 3 B; Azorubin; Carmoisin B.

Scientific name.—Sodium salt of para-sulphonaphthalene-azo-alpha-naphthol-para-sulphonic acid.

Discovered and patented.—1883.

Shade.—Red. Offered by 6 out of 12 sources.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Cazeneuve and Lépine: Not poisonous to human beings.
- 3. Meyer (J. Amer. Chem. Soc. 1907, v. 29, p. 898): One hundred milligrams per kilogram body weight, or 70 grains per 100 pounds body weight administered, increased geometrically; diarrhea was marked only after administration of exceptionally large doses (the seventh day); the stools were deep violet and the urine was carmine, becoming dark; the autopsy developed nothing abnormal; the whole interior was of a red color.

4. LIEBER (p. 138): A guinea pig received once a day six times every other day 241 milligrams per kilogram body weight, or 169 grains per 100 pounds body weight; appetite remained good throughout, and aside from an occasional thirstiness noted no untoward observations were recorded.

## G. T. 105.

Trade names.—Fast Red E; Fast Red.

Names under which it was offered on the United States market as a food color in 1907.—Claret Red RZ.

Scientific name.—Sodium salt of para-sulphonaphthalene-azo-beta-naphthol-monosulphonic acid.

Discovered and patented.—1878.

Shade.—Red. Offered by 1 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Meyer (J. Amer. Chem. Soc. 1907, v. 29, p. 898): Initial dose 100 milligrams per kilogram body weight or 70 grains per 100 pounds, and given three times increased geometrically once before diarrhea was observed; there was no albumen in the urine; urine was colored deep red; feces colored red at the start, at the end of the experiment chocolate brown; autopsy showed all parts to be substantially normal.
- CAZENEUVE AND LÉPINE (Compt. rend., 1885, v. 101, pp. 823-827): A. Dog: Weight 21.5 kilo, received as follows:

Days.	Grams.	Milligrams per kilo.	Grains per 100 pounds.
32	0. 500	23. 2	16. 2
20	2. 150	100. 0	70. 0
8	4. 300	200. 0	140. 0
10	5. 000	232. 0	162. 0
5	10. 000	462. 0	324. 0

Nothing abnormal except occasional greenish urine; no vomiting; no diarrhea. B. Man: 1. One of the experimenters took 1 gram, dissolved in wine, daily for 15 days; no effect. 2. A man aged 25, afflicted with albuminuria, received the following:

Days.	Grams.
3	0. 5
2	1. 0
1	2. 0

Caused colic without diarrhea; amount of urine or albumen not affected. 3. Three men afflicted with Bright's disease received each daily for 8 days 1 gram; no effect on the albumen. 4. A 30-year-old man chronic invalid took 4 grams one day, 6 grams the next day; no effect was observed.

4. Arloing and Cazeneuve (Archives de physiologie, 1887, pp. 356-393): As the result of this work, which is divided into four parts: (1) Stating the effect of direct introduction of the color into the circulation; (2) intravenous injections; (3) comparing the effects of injections of color and of salt; and (4) feeding by the mouth—these investigators conclude that these coloring matters are toxic only in extremely large doses; that when given to dogs with their food no inconvenience of any kind results; this is based upon experiments upon three dogs, covering 145 days, where each dog received, per kilogram initial body weight, in the first case, 20,307 milligrams, or 14,215 grains per 100 pounds initial body weight; in the second case, 29,590 milligrams, or 20,713 grains per 100 pounds initial body weight. Per day this would mean 98 grains per 100 pounds initial body weight in the first case; in the second case the daily dose was 143 grains per 100 pounds initial body weight.

## G. T. 106.

Trade names.—New Coccin; Cochineal Red A; Brilliant Scarlet; Crocein Scarlet 4 BX.

Names under which it was offered on the United States market as a food color in 1907.—Claret Red RZ; Crocein Scarlet; New Coccin; Scarlet L; Brilliant Scarlet 4 R.

Scientific name.—Sodium salt of para-sulphonaphthalene-azo-beta-naphthol-disulphonic acid (G.).

Discovered.—1878. Offered by 5 out of 12 sources.

Shade.—Red.

### FAVORABLE.

1. CAZENEUVE AND LÉPINE. (See Weyl, p. 115.)

2. Weyl (p. 31): "\* \* \* not poisonous to human beings and dogs, \* \* \* Purple \* \* \* "

3. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd., 1886, p. 643): Tolerated by man, sick or well.

### UNFAVORABLE.

1. Prohibited by Confectioners' List.

### G. T. 107.

Trade names.—Fast Red D; Azo Acid Rubin 2 B; Fast Red E B; Bordeaux S; Amaranth.

Names under which it was offered on the United States market as a food color in 1907.—Claret Red RZ; Red; Amaranth B (Azo color similar to); Bordeaux S; Naphthol Red S; Amaranth.

Scientific name.—Sodium salt of para-sulphonaphthalene-azo-beta-naphthol-disulphonic acid (R).

Discovered and patented.—1878.

Shade.—Red. Offered by 7 out of 12 sources.

## FAVORABLE.

- 1. CAZENEUVE AND LÉPINE (see Weyl, p. 115): Not poisonous to human beings.
- 3. Lieber (p. 148): A rabbit received 284 milligrams per kilogram body weight, or 199 grains per 100 pounds once a day, five times every other day, and aside from decreased appetite the second and third day of the observation period nothing untoward is noted.
- 4 Weyl (p. 31): "\* \* \* not poisonous to human beings and dogs, \* \* \* Purple \* \* \*"
- CAZENEUVE AND LÉPINE (Bull de l'acad. de méd., 1886, p. 643): Tolerated by man, sick or well.

## G. T. 138.

Trade names.—Fast Brown G; Acid Brown.

Scientific name.—Sodium salt of bi-sulphobenzene-disazo-alphanaphthol.

Discovered.—1882.

Shade.—Brown. Not offered.

### FAVORABLE.

1. Weyl (p. 134): "The following is a summary of the results obtained with the Disazo colors submitted to test by me, viz, Fast Brown G \* \* \*. All these proved to be nonpoisonous \* \* \*." A. A dog weighing 9.63 kilos received 312 milligrams per kilogram body weight, or 218 grains per 100 pounds body weight; the same dose was repeated 48 hours afterwards, when diarrhea set in. and the urine was colored red; 24 hours afterwards 208 milligrams per kilogram body weight, or 146 grains per 100 pounds body weight were given; the urine was colored strongly red 24 hours thereafter. Eight days afterwards 520 milligrams per kilogram body weight, or 364 grains per 100 pounds body weight, were administered; marked diarrhea set in; 3 days later the dose given was twice the dose last given, when severe diarrhea resulted, but unchanged color appeared in the urine, and continued for 24 hours afterwards; diarrhea continued for 96 hours. Evidences of albuminuria apparently not dependable. B. A second dog, weighing 5.9 kilos, received 339 milligrams per kilogram body weight, or 237 grains per 100 pounds body weight daily throughout an entire month; diarrhea was produced after 6 days, which continued almost during the month; appetite was diminished and the loss in weight was about 20 per cent on the original. C. Weyl concludes as follows: "According to these experiments, this color in continuous, though slight, doses, or in large doses, but less frequently, produces diarrhea, anorexia, and emaciation." (p. 136.)

#### UNFAVORABLE.

1. Prohibited by Confectioners' List.

2. Weyl (p. 147): "Of the remaining colors \* \* \* others (produce) diarrhea (Fast Brown \* \* \*)."

## G. T. 160.

Trade name.—Crocein Scarlet 3 B; Ponceau 4 RB.

Scientific name.—Sodium salt of sulphobenzene-azo-beta-naphthol-monosulphonic acid.

Discovered and patented.—1881.

Shade.—Scarlet. Not offered.

FAVORABLE.

Nothing.

UNFAVORABLE.

1. Prohibited by Confectioners' List.

DOUBTFUL.

1. Houghton (J. Amer. Chem. Soc., 1907, v. 29, pp. 1351-57): Hinders fibrin digestion at all strengths; at 1:200 hinders casein and albumen digestion.

## G. T. 163.

Trade names.—Biebrich Scarlet; Ponceau B; New Red L; Ponceau 3 RB; Fast Ponceau B; Imperial Scarlet.

Scientific name.—Sodium salt of sulphobenzene-azo-sulpho-benzene-azo-beta-naphthol.

Discovered.—1878.

Shade.—Scarlet. Not offered.

FAVORABLE.

1. Permitted by law in Italy. (See Lieber, pp. 18, 22, 23.)

UNFAVORABLE.

1. Prohibited by Confectioners' List.

## G. T. 164.

Trade name.—Crocein Scarlet O extra.

Scientific name.—Sodium salt of sulphobenzene-azo-sulphobenzene-azo-beta-naphthol-sulphonic acid.

Discovered.—1888.

Shade.—Scarlet. Not offered.

FAVORABLE.

Nothing.

UNFAVORABLE.

1. Prohibited by Confectioners' List.

## G. T. 166.

Trade name.—Wool Black.

Scientific name.—Sodium salt of sulphobenzene-azo-sulphobenzene-azo-para-tolyl-beta-naphthylamin.

Discovered.—1885.

Shade.—Black. Not offered.

### FAVORABLE.

1. Permitted by Confectioners' List.

2. Weyl (p. 134): "The following is a summary of the results obtained with the Disazo colors submitted to test by me, viz: \* \* \* Wool Black \* \* \*.

All these proved to be nonpoisonous \* \* \*."

3. A. A dog received 167 milligrams per kilogram body weight, or 117 grains per 100 pounds body weight each day for 3 successive days; no color was administered for the next 2 days, and the third day the dose was double the former dose. The urine was colored bluish-black and contained albumen. Two days afterward the same dose was given; urine of intense dark-blue color, and contained unaltered coloring matter, which disappeared in 48 hours. Albuminuria continued for about 15 days. (p. 137.) B. "Wool Black is nonpoisonous both by gastric and by subcutaneous administration." (p. 137.)

## G. T. 169.

Trade names.—Crocein Scarlet 7 B; Ponceau 6 RB; Crocein Scarlet 8 B.

Names under which it was offered on the United States market as a food-color in 1907.—Sodium salt of sulphotoluene-azo-beta-naphthol-alpha-sulphonic acid.

Discovered and patented.—1881.

Shade.—Red. Offered by 1 out of 12 sources.

### FAVORABLE.

1. Permitted by Confectioners' List.

## G. T. 188.

Trade names.—Naphthol Black B; Brilliant Black B.

Name under which it was offered on the United States market as a food-color in 1907.—Naphthol Black BDF.

Scientific name.—Sodium salt of disulpho-beta-naphthalene-azo-alpha-naphthalene-azo-beta-naphthol-disulphonic acid.

Discovered and patented.—1885.

Shade.—Black. Offered by 1 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 138): A. A dog received 112 milligrams per kilogram body weight, or 78 grains per 100 pounds body weight, daily for 3 successive days; distinct albuminuria, uncolored urine; blue-colored feces; thereupon the daily dose was increased to 187 milligrams per kilogram body weight, that is, 131 grains per 100 pounds. No color was administered for 6 days, and during this time the urine was colored from reddish-violet to a bluish-black red; thereupon the last dose was doubled and 24 hours afterwards that dose was doubled; rather much albumen in urine which was bluish; albuminuria continued for about a week. B. A dog received 222 milligrams per kilogram body weight, or 155 grains per 106 pounds daily throughout a whole month; it remained entirely well with good appetite.
- 3. "This color is harmless when administered by the stomach, but poisonous subcutaneously." (p. 140.)
- 4. Schacherl: Not harmful under conditions in which it is used.

### UNFAVORABLE.

1 Weyl (p. 139): Where a dog, receiving subcutaneously 31 milligrams per kilogram body weight, or 22 grains per 100 pounds body weight, subcutaneously, died apparently wholly as a result of the color.

2. "Naphthol Black P, however, is plainly poisonous when introduced into the subcutaneous cellular tissue." (p. 147.)

## G. T. 197.

Trade names.—Bismarck Brown; Phenylene Brown; Leather Brown; English Brown; Manchester Brown; Vesuvin; Cinnamon Brown.

Names under which it was offered on the United States market as a food-color in 1907.—Vesuvin 4B Conc. Z; Bismarck Brown; Bismarck Brown B No. 216; Bismarck Brown B.

Scientific name.—Hydrochlorid of benzene-disazo-phenylene-diamin, Discovered and patented.—1863.

Shade.—Reddish brown. Offered by 4 out of 12 sources.

### FAVORABLE.

1. Weyl (p. 115): "Nonpoisonous Bismarck Brown."

### UNFAVORABLE.

- 1. Prohibited by Confectioners' List.
- 2. Weyl (p. 117): A. A dog received 33 milligrams per kilogram body weight, or 23 grains per hundred pounds body weight; in one and one-half hours, vomiting; next day same dose same result with the addition that the animal took no food and moved about but little for 48 hours; on the fourth day same dose, in two hours, vomiting; for 96 hours animal took no food; on the eighth day albumen in the urine and the animal ate; on the ninth day 83 milligrams per kilogram body weight or 58 grains per 100 pounds body weight, vomited for one-half hour after administration; for the next 5 days the animal took hardly any food; on the fourteenth day the animal improved, took food on the fifteenth day; traces of albumen in urine for 17 days longer, at end of which time animal recovered.
  - B. A dog received 169 milligrams per kilogram body weight, or 118 grains per 100 pounds body weight; in 24 hours the urine was colored brown; 48 hours after the first dose that dose was repeated, and unconverted color was found in the urine; 48 hours later the same dose was repeated; the color in the urine disappeared in 24 hours; 4 days later the dose was trebled, and the animal vomited, seemed sick for 4 days thereafter; on the fifth day recovery apparently complete. On the sixth day the last dose was repeated, and the animal vomited after the administration. It took no food for 24 hours, and was normal after 48 hours. There was no albuminuria in this case.
  - C. A dog received daily for an entire month 45 milligrams per kilogram body weight, or 31½ grains per 100 pounds body weight; it was in good health during the entire time, did not vomit, and ate as usual. Its gain in weight was 6½ per cent.
- 3. "Bismarck Brown produces, when administered to dogs by the stomach, even in doses of 350 milligrams per kilogram body weight (245 grains per 100 pounds body weight), vomiting and albuminuria. Further disturbance is not noted even in large doses; small doses, 45 milligrams per kilogram body weight (or 31½ grains per 100 pounds body weight), even when frequently administered seem to be entirely harmless. Doses of 16 milligrams are harmless even when introduced into the subcutaneous cellular tissue." (p. 118.)
- 4. "Of the remaining colors some produce vomiting (e. g., Bismarck Brown \* \* \*)." (p. 147.)

5. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): Produces eczema, and cites

Deutsch Med. Wochenschr., 1891, p. 45.

6. Fraenkel (p. 575): "When the Azo dyestuffs do not contain any sulpho group they are poisonous. Thus, for example, Bismarck Brown \* \* \*. This produces no effect in small doses; on the other hand, doses of 350 milligrams per kilogram of animal (245 grains per 100 pounds) produce albuminuria and vomiting."

#### DOUBTFUL.

HOUGHTON (J. Amer. Chem. Soc., 1907, v. 29, pp. 1351-1357): Hinders digestion
of fibrin, casein and albumen, in strengths of 1:100 or 1:400.

## G. T. 201.

Trade names.—Manchester Brown EE; Bismarck Brown R; Vesuvin B.

Names under which it was offered on the United States market as a food color in 1907.—Vesuvin 4 B Conc. Z; Vesuvin B.

Scientific name.—Hydrochlorid of toluene-disazo-meta-tolylene-diamin.

Discovered and patented.—1878.

Shade.—Reddish brown. Offered by 2 out of 12 sources.

### UNFAVORABLE.

 Lewin (Lehrbuch der Toxicologie, 1897, p. 231): "Produces eczema," and cites Deutsch. Med. Wochenschr., 1891, p. 45.

Note.—The literature is not always conclusive as between Nos. 197 and 201, and probably most, if not all, the references under No. 197 also apply to No. 201.

## G. T. 240.

Trade name.—Congo Red.

Name under which it was offered on the United States market as a food color in 1907.—Congo.

Scientific name.—Sodium salt of diphenyl-disazo-binaphthionic

acid.

Discovered and patented.—1884.

Shade.—Red. Offered by 1 out of 12 sources.

### FAVORABLE.

1. WEYL: "According to Experiments 1 and 2 Congo Red is, after long-continued administration by the stomach, harmless."

2. A. A dog received 274 milligrams per kilogram body weight, or 192 grains per 100 pounds; this dose was repeated next day, when the urine became pale, was strongly alkaline and contained albumen; the next day the same dose was repeated, whereupon the urine was of a weak red color, and a little albumen present; the next day the dose was increased to 411 milligrams per kilogram body weight, or 298 grains per 100 pounds; the urine was somewhat reddish and contained a little albumen. No color was given for 6 days, at the end of which time the urine was reddish, and deposited a reddish sediment, probably Congo. At the end of that time the dose was increased to 685 milligrams per kilogram

### 2. A.—Continued.

body weight, or 480 grains per 100 pounds body weight; the urine was feebly alkaline and contained some albumen. The next day the dose last given was doubled; the urine was colored to such an extent that it could be dyed with. Two days later the same dose was repeated; animal took but little food, was otherwise comfortable; there was little albumen present. The loss in body weight was about 4½ per cent. (p. 141.)

B. A second dog received 233 milligrams per kilogram body weight, or 163 grains

per 100 pounds, daily for one month, and remained entirely well.

## G. T. 269.

Trade name.—Chrysamin R.

Name under which it was offered on the United States market as a food color in 1907.—Chrysamin R.

Scientific name.—Sodium salt of ditolyl-disazo-bisalicylic acid.

Discovered and patented.—1884.

Shade.—Yellow. Offered by 1 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 134): "The following is a summary of the results obtained with Disazo colors submitted to test by me, viz: Chrysamin R." "All of these proved to be nonpoisonous, \* \* \*."
- 3. A. A dog received 515 milligrams per kilogram body weight, or 361 grains per 100 pounds; urine became alkaline and yellowish, and easily dyed cotton; very little albumen. Within 24 hours the same dose was repeated, diarrhea resulting and vomiting for 3 days, whereupon the animal was given one-fifth of the dose, or 103 milligrams per kilogram body weight, that is, 72 grains per 100 pounds body weight; the urine continued slightly colored, and contained a distinct amount of albumen, continuing for 2 days, when the last dose was repeated; 24 hours afterwards the dose was doubled, and repeated the next day; the day after the dose was increased 50 per cent; 3 days later the last dose was repeated; slight albuminuria set in, lasting 5 days. B. A dog received three doses of 619 milligrams each per kilogram body weight, or 433 grains per 100 pounds body weight, three times in the course of 10 days; the urine was yellowish in color and contained very little albumen. (p. 145.)
- 4. "Chrysamin is harmless when taken into the stomach." (p. 147.)
- 5. Schacherl (p. 1045) says Chrysamin is harmless under the conditions in which it is used.

### UNFAVORABLE.

1. Weyl (p. 147): "Of the remaining colors some produced vomiting \* \* \* others diarrhea (\* \* \* Chrysamin R \* \* \*.'')

## G. T. 277.

Trade names.—Benzopurpurin 4 B; Cotton Red 4 B; Sultan Red 4 B.

Scientific name.—Sodium salt of ditolyl-disazo-binaphthionic acid. Discovered.—1884-85.

Shade.—Red. Not offered.

#### FAVORABLE.

Nothing.

### UNFAVORABLE.

Chlopin (p. 130): On his own experiments classes it as suspicious. The experimental data are as follows:

### Experimental data by Chlopin.

[1 gram=125 mg=87.5 grains.]

Date.	Dose.	Weight.	24-hour urine.	General condition of animal and urine.
1901. Apr. 16	Grams.	Kilos.	cc. 300 330	Dog is well, lively; urine normal color; acid; no albumen.
18 19 20	3		450 470 370 320 310 370 310	Vomiting several times; no albumen. Urine yellow-orange; no albumen; no vomiting. Do.
21 22	2			Do. Do.
23 24				Urine normal; dog is well. Do.
Total.	7			

### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm. 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 287.

Trade name.—Azo Blue.

Name under which it was offered on the United States market as a food color in 1907.—Azo Blue.

Scientific name.—Sodium salt of ditolyl-disazo-bi-alpha-naphthol-para-sulphonic acid.

Discovered.—1885.

Shade.—Grayish violet. Offered by 1 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 134): The following is a summary of the results obtained with the Disazo colors submitted to test by me, viz: \* \* \* Azo-blue, \* \* \* .'' "All these proved to be nonpoisonous."
- 3. "Azo-blue is harmless, both when administered by the stomach and subcutaneously." A. A dog received 237 milligrams per kilogram body weight, or 166 grains per 100 pounds; 2 days later this same dose was repeated, and the urine was a violet color strongly alkaline and contained a little albumen. The next day the dose was increased to 2.5 times; colorless urine, and little albumen; there was no phenol. The next day 4 times the original dose was administered; abundant bluish-violet urine and little albumen. No administration for 4 days, when 2.5 times the original dose were given, and that dose repeated 3 days later; during this time the urine was colorless and contained little albumen; there was a very slight increase in weight. B. A dog received 319 milligrams per kilogram body weight, or 233 grains per 100 pounds daily for one month; animal remained well with good appetite; a slight amount of albumen made its appearance in the urine. (p. 144.)

#### UNFAVORABLE.

1. Weyl (p. 144): "A slight amount of albumen made its appearance in the urine."

## G. T. 394.

Trade names.—Dinitrosoresorcin; Dark Green; Russian Green; Alsace Green; Fast Green O; Chlorin; Fast Myrtle Green.

Scientific name.—Dinitroso-resorcinol (Dioximidoquinone).

Discovered and patented.—1875.

Shade.—Green. Not offered.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 63): "According to the above experiments Dinitroso-resorcinol is not dangerous to dogs when administered by the stomach even in large doses; while hypodermic administration proves fatal within 24 hours, in the proportion of 190 milligrams per kilogram of body weight" (that is, 132 grains per 100 pounds).
- 3. A. A dog received 173 milligrams per kilogram body weight, or 121 grains per 100 pounds; scanty dark-brown urine; the next day the same dose was repeated and urine continued dark brown, and contained trace of albumen as well as distinct reaction for iron; the next day the dose was repeated; animal remained lively; the day after that the dose was increased 50 per cent; no albumen. The loss in body weight was only 4 per cent. B. A dog received 198 milligrams per kilogram body weight, or 139 grains per 100 pounds; seems to have been loss of appetite, coupled with dark-brown, almost black urine; 2 days later the dose was doubled, and the urine was colored green by ferrous oxid, and contained no albumen nor sugar; 2 days later the dose was increased 50 per cent, and some albumen was then found in the urine. (p. 62.)

## G. T. 398.

Trade name.—Naphthol Green B.

Names under which it was offered on the United States market as a food color in 1907.—Naphthol Green; Naphthol Green B.

Scientific name.—Ferrous sodium salt of nitroso-betanaphthol-beta-monosulphonic acid.

Discovered.—1883.

Shade.—Green. Offered by 2 out of 12 sources.

### FAVORABLE.

1. Weyl (p. 64): A. A dog received 172 milligrams per kilogram body weight, or 120 grains per 100 pounds body-weight; the urine was greenish, and conjunctiva stained intensely green; dose was repeated 3 successive days, and the next day the dose was increased to fivefold; appetite undisturbed, and animal remained lively. B. A dog received 417 milligrams per kilogram body weight, or 292 grains per 100 pounds; there was a dirty yellowish-green color to the urine; no albumen, and not more than traces of iron. The animal was normal within a day, and two days after the same dose was repeated; no change in animal was recorded, except that on the day following the feces were normal, but colored green.

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- 2. Weyl (p. 65): "Experiments 1 and 2, in which 2 to 5 grams of the color were introduced directly into the stomach, demonstrated its harmlessness in this method of administration."
- 3. Buss lists it as nonpoisonous.

### UNFAVORABLE.

1. Prohibited by Confectioners' List.

 Weyl (p. 65): "On the other hand, in a hypodermic administration, in two out of three cases abscesses and septic fever were induced."

## G. T. 399.

Trade names.—Sun Yellow; Curcumin S; Jaune Soleil; Maize. Scientific name.—Sodium salt of the so-called Azoxy-stilbene-disulphonic acid.

Discovered.—1883.

Shade.—Yellow. Not offered.

### FAVORABLE.

1. Meyer (J. Amer. Chem. Soc., 1907, v. 29, p. 897): A dog received 100 milligrams per kilogram body weight, or 70 grains per 100 pounds, increased geometrically through the fourth day, when diarrhea set in; up to this time the animal had been given 19.27 grams, or 1,465 milligrams per kilogram body weight, equivalent to 1,026 grains per 100 pounds body weight; the average dose per day would have been 366 milligrams per kilogram body weight, or 256 grains per 100 pounds; the animal was given its fifth portion of coloring matter the same size as the fourth, thereupon color was omitted, and for the following 7 days the dosage of the third day, which amounted to 400 milligrams per kilogram body weight, or 280 grains per 100 pounds body weight, was given; the urine was colored orange throughout the entire test after the first day; the fecal matter also was of orange color; slight diarrhea on the fourth and twelfth days of the test, and vomiting on the fifth day, the cause of which does not seem to have been definitely determined; the autopsy revealed nothing abnormal.

### G. T. 425.

Trade names.—Auramin; Auramin O; Pyoctanin Aureum.

Names under which it was offered on the United States market as a food color in 1907.—Auramin O; Auramin; Canary Yellow.

Scientific name.—Hydrochlorid of imido-tetramethyl-diamido-diphenylmethane.

Discovered.—1883.

Shade.—Greenish yellow. Offered by 3 out of 12 sources.

FAVORABLE.

Nothing.

### UNFAVORABLE.

Chlopin (p. 157): On his own experiments classes it as poisonous. The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram=69 mg=48 grains.]

Date.	Dose.	Weight.	24-hours' urine.	General condition of animal and urine.
1901. Mar. 2-4 6 7	Grams.	Kilos. 14.4	cc. 550 340	Dog quite normal; acid; no albumen. Urine strong brown yellow; traces of albumen; appetite less. Thin stool.
8 9 10				Diarrhea; urine brown yellow, pales with sulphuric acid, and contains much albumen.  Stool normal; color of urine weaker; much albumen; eats well; lively.  Stool normal; urine less colored; less albumen.
11 12 13			335   Stool normal; urine less colored; trac	Stool normal; urine less colored; traces of albumen. Stool normal; urine less colored; insignificant traces of albumen.
14 15	2.0	13. 2		Do. Strongly brown yellow; much albumen.
16 17			None. 295	Dog depressed; lies down; eats little.  Vomits and diarrhea; urine strong yellow; little albumen; takes only milk.
19 20			480	Vomits and diarrhea; urine strong yellow; much albumen. Feces normal color; color urine almost normal; much albumen.
21 24	1 0. 4	13. 6	352 380	Feces normal color; color urine almost normal; albumen less. Do.

<sup>&</sup>lt;sup>1</sup> Subcutaneously.

### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 427.

Trade names.—Malachite Green; New Green; Fast Green; Benzal Green; Diamond Green B; Malachite Green B; New Victoria Green; Vert Diamant; Bitter-almond-oil Green.

Names under which it was offered on the United States market as a food color in 1907.—Green M; New Green Crystals; Green 088.

Scientific name.—Zinc double-chlorid, oxalate, ferric double-chlorid of tetramethyldi-para-amido-triphenyl-carbinol.

Discovered.—1877-78.

Shade.—Bluish green. Offered by 2 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- Weyl (p. 24): "According to Grandhomme \* \* \* Malachite Green are (is) also nonpoisonous."
- 3. "\* \* \* Malachite Green are (is) as is now established, almost without poisonous action." (p. 55.)
- Lewin (Lehrbuch der Toxikologie, 1897, p. 231) says when free from arsenic it is harmless.
- 5. Buss lists it as nonpoisonous.

### UNFAVORABLE.

1. Penzoldt (Archiv. exper. path. pharm., 1890, v. 26, p. 312): One hundred milligrams per kilogram body weight of rabbit, or 70 grains per 100 pounds, injected subcutaneously, caused after the third day motor paralysis and occasional cramps, which resulted fatally at the end of the ninth day.

2. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): In the case of one workman, in contrast with others who had long been unaffected by this substance, itching, burning, inflammation, and swelling of hands and feet, and formation of blisters occurred.

## G. T. 428.

Trade names.—Brilliant Green; New Victoria Green; Emerald Green; Malachite Green B; Ethyl Green; Fast Green J.

Names under which it was offered on the United States market as a food color in 1907.—Green E; Green 087; Emerald Green Crystals.

Scientific name.—Sulphate of zinc-double-chlorid (rarely oxalate) of tetraethyl-diamido-triphenyl-carbinol.

Discovered.—1879-80.

Shade.—Yellowish green. Offered by 3 out of 12 sources.

### FAVORABLE.

- Lewin (Lehrbuch der Toxikologie, 1897, p. 231) says when free from arsenic it is harmless.
- 2. Buss lists it as nonpoisonous.

#### UNFAVORABLE.

1. Chlopin (pp. 171-2): Classifies it as "very poisonous" on his own experiments (see p. 181). The experimental data are as follows:

## Experimental data by Chlopin.

No. 1. [1 gram=125 mg=87.5 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 24 25	2	Kilos. 8	cc. 410	Before experiment dog is well; urine normal. Soon after giving dye vomiting and diarrhea; in the evening only drank water; vomiting kept up. No vomiting; urine greenish; acid; insignificant traces of albumen. Do.
28 29 30 May 1	2		395 395	No vomiting; urine greenish; acid; no albumen. Do. Do. Urine normal; dog quite well. Do. Do.
Total	. 4			

No. 2.

### (1 gram=111 mg=78 grains.)

1902. Jan. 21 23–24 25	3	9 7. 2	400	Before experiment dog quite normal; urine normal; after giving dye very violent vomiting several times. Urine greenish in color; acid; very much albumen; no vomiting. Do.
26 Total	5			Died during the night.

### Experimental data by Chlopin—Continued.

No. 3.

## [1 gram=119 mg=83 grains.]

Date.	Dose.	Weight	24 hours' urine.	General condition of animal and urine.
1902. Jan. 28	Grams.	Kilos. 8. 4	cc.	Before experiment dog and urine normal; soon after giving dve
Jan. 28	3	8.4	370	vomiting began, lasting over an hour; dog stands with
29	2			difficulty.  During night dog improved somewhat, began to eat; drinks
30				much; soon after giving dye vomiting.  During night vomiting and darphea; during night 30th, 31st,
31				in bad condition; does not take food.  During night 31st second dog found dead in cage. Cause of death, "paralysis of the heart."
				death, "paratysis of the heart."
Total	5			

2. Lewin (*Lehrbuch der Toxikologie*, 1897, p. 231): In the case of one workman, in contrast with others who had long been unaffected by this substance, itching, burning, inflammation and swelling of hands and feet, and formation of blisters occurred.

## G. T. 433.

Trade name.—Guinea Green.

Name under which it was offered on the United States market as a food color in 1907.—Guinea Green B.

Scientific name.—Sodium salt of diethyldibenzyl-diamido-triphenyl-carbinol-disulphonic acid.

Discovered.—1883.

Shade,—Green. Offered by 1 out of 12 sources.

### FAVORABLE.

Chlopin (p. 174): On his own experiments classified it as "nonpoisonous." The
experimental data are as follows:

Experimental data by Chlopin.

### [1 gram=200 mg=140 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 18 19 20 21 222 23 24 25 26 27 28 30 31 Total.	2 2 2 2 10	Kilos.	250 235 280 290 270 270 260 240 290 246 243 233 233	Dog and urine normal before experiment. Urine slightly greenish, no albumen; vomited at night. No vomiting. Urine slightly greenish; no albumen. Urine normal in color and composition. Do. Do. Do. Do. Do. Do. Do. Do. Do. Do

## G. T. 434.

Trade names.—Light Green SF bluish; Acid Green.

Scientific name.—Sodium salt of dimethyldibenzyl-diamido-triphenyl-carbinol-trisulphonic acid.

Discovered.—1879.

Shade.—Green. Offered by 1 out of 12 sources.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (pp. 176-7): Examined this color, and on his own experiments classes it as "nonpoisonous, but not entirely indifferent." The experimental data are as follows:

Experimental data by Chlopin.

No. 1.

### [1 gram=133 mg=93 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Apr. 24 25 26 27 28	Grams.	Kilos. 7.5	cc. 380 347 297	Dog and urine normal. Urine green; acid; no albumen. Urine green; traces of albumen. Do. Do.
29 30 31 May 1-3	4		360	Urine green; no albumen. Urine less green; no albumen. Do. Do. Do. Do.

### No. 2.

### [1 gram=110 mg=77 grains.]

1902. Aug. 5 6 7 8 9 10 11 12 Total .	3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3	9.1	300 310 320 350 400 350	Dog and urine normal. Urine greenish, acid, no albumen. Do. Do. Do. Do. Do. Do. Do. Do.	
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### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm. 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 435.

Trade names.—Light Green SF yellowish; Acid Green; Acid Green extra conc.

Names under which it was offered on the United States market as a food color in 1907.—Acid Green conc. V N; Light Green SF yellow shade; Acid Green conc. 780; Pistachio.

Scientific name.—Sodium salt of diethyldibenzyl-diamido-triphenyl-carbinol-trisulphonic acid.

Discovered.—1879.

Shade.—Green. Offered by 4 out of 12 sources.

### FAVORABLE.

1. Lieber (p. 144): The animal was a fully developed male guinea pig, and received per kilogram body weight 240 milligrams, or 168 grains per 100 pounds, five times in all, every other day. There was apparently nothing irregular or abnormal observed during the whole test of nine days.

## G. T. 448.

Trade names.—Magenta; Fuchsin; Rosein; Anilin Red.

Obsolete names.—Rubin; Solferino; Fuchsiacin; Rubianite; Azalein; Erythrobenzin; Harmalin.

Names under which it was offered on the United States market as a food color in 1907.—Magenta powder A; Fuchsin Crystals; Magenta F A B S Red 101.

Scientific name.—Mixture of hydrochlorid or acetate of pararosanilin (triamidotriphenylcarbinol) and rosanilin (triamidodiphenyltolylcarbinol).

Shade.—Bluish-red. Offered by 4 out of 12 sources.

### FAVORABLE.

1. Permitted by Confectioners' List.

2. Weyl (p. 22): "The colors examined \* \* \* Fuchsin were (was) found to be nonpoisonous;" "Similarly a hen which had eaten for three weeks oats covered with fuchsin was in good health." (p. 24.)

3. "According to Grandhomme rabbits bear without injury fuchsin free from arsenic \* \* \*." (p. 31.)

- 4. "Fuchsin \* \* \* (is) as is now established, almost without poisonous action." (p. 55.)
- 5. Fraenkel (p. 574), quoting Penzoldt, says that it is entirely nonpoisonous, and completely prevents putrefaction.

6. Permitted by the law of Austria.

- 7. Lewin (Lehrbuch der Toxikologie, 1897, p. 230), says when free from arsenic it is harmless.
- 8. CLOUET AND BERGERON (J. pharm. chim., 1871, v. 25, p. 296): One of them took personally 500 milligrams, that is, 7.7 grains in 16 days; there was no digestive disturbance of any kind, and the urine, which was examined daily, contained no albumen. They cite a case of Bright's disease, in which the amount of albumen decreased when fuchsin was administered, and they conclude that fuchsin may be good for sufferers from Bright's disease.

### UNFAVORABLE.

1. Forbidden by the law of France. (See Lieber, p. 31.)

2. Chlopin (p. 178): Examined this color, and on his own experiments classes it as "Suspicious because of vomiting and traces of albumen." The experimental data are as follows:

### Experimental data by Chlopin.

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 18 19 20 21 22 23 24 Total.	Grams. 3 2 5	Kilos.	270 280 320	Dog before experiment well and urine normal; vomited several times after receiving dye. Urine quite red; acid; no albumen; general condition normal. Do. Urine less colored; acid; no albumen; vomited once. Urine darkish; acid; traces albumen. Color normal; no albumen. Do.

Note.—This sample may have contained some phosphin, G. T. 532, see page 133.

#### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm. 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 450.

Trade names.—Hofmann Violet; Dahlia; Red Violet 5R extra; Violet R; Iodin Violet; Primula; Violet 5 R; Violet R R.

Scientific name.—Mixture of the hydrochlorids or acetates of the monodi- or trimethyl- (or ethyl-) rosanilins and pararosanilins.

Discovered.—1863.

Shade.—Violet. Not offered.

### FAVORABLE.

- 1. Weyl (p. 24): "\* \* \* Anilin Violet (Dahlia) \* \* \* (is) also nonpoisonous."
- 2. Buss lists it as nonpoisonous.

### UNFAVORABLE.

 FRAENKEL (p. 574) quotes Penzoldt, and says it completely arrests development, and causes muscular paralysis.

## G. T. 451.

Trade names.—Methyl Violet B; Direct Violet; Dahlia; Paris Violet; Violet de Methylanilin; Pyoctanin.

Names under which it was offered on the United States market as a food color in 1907.—Methyl Violet; Methyl Violet B; Methyl Violet BB extra; Methyl Violet 3 B D.

Scientific name.—Hydrochlorid of penta- and hexamethyl- para-rosanilin.

Discovered.—1861.

Shade.—Violet. Offered by 5 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Permitted by the Austrian law.

- 3. Weyl (p. 24): "\* \* \* Anilin Violet (Dahlia) \* \* \* [is] also non-poisonous."
- poisonous."

  4. "\* \* Methyl Violet [is] as is now established, without poisonous action."

  (p. 55).
- 5. Fraenkel (p. 573): "Methyl Violet \* \* \* is relatively nonpoisonous."
- 6. Buss lists it as nonpoisonous.

#### UNFAVORABLE.

- 1. Graefe and Braunschweig (Fortschr. Medizin, 1890, v. 8, p. 405): "It seems to be proven that damage will actually result even in the case of most cautious use, which we are sure we exercised."
- 2. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 52): I. A dog weighing 7,600 grams received 5.6 grams dye in 12 days; this amounts to 61 milligrams per kilo per day, or 43 grains per 100 pounds per day. Continued vomiting beginning with 0.1 gram dye; progressive emaciation and general falling away; catarrh of eyes and nose; distinct dislike for food and great desire to sleep; temperature below normal, urine unchanged. Loss of weight 1,600 grams, or 21 per cent. The animal died on the thirteenth day. The autopsy showed a pale and blood-poor liver; the kidneys were in a typically congested condition and contained accumulations of blood corpuscles. II. A second dog weighing 6,000 grams received 3.8 grams dye in 14 days, which amounts to 71.4 milligrams per kilo per day, or 50 grains per 100 pounds per day. The animal died on the fourteenth day. There was daily vomiting and rapid emaciation; final weight loss was 1,100 grams, or 18.3 per cent; temperature normal; bloody urine beginning the eighth day. The autopsy showed a blood-poor liver, soft and swollen epithelia; kidneys the same as in the case of the preceding dog.

## G. T. 457.

Trade names.—Anilin Blue, spirit-soluble; Spirit Blue; Fine Blue; Bleu Lumière; Opal Blue; Gentian Blue 6B; Hessian Blue; Bleu-de-Nuit.

Scientific name.—Hydrochlorid sulphate or acetate of triphenylrosanilin and triphenylpararosanilin.

Discovered.—1860-1862.

Shade.—Greenish blue; not offered.

### FAVORABLE.

1. Permitted by Confectioners' List.

- 2. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 48) says it is harmless. A dog weighing 4,500 grams received 17 grams dye in 30 days, which amounts to 126 milligrams per kilo per day or 88 grains per 100 pounds per day. Weight remained the same, general condition good, urine and temperature unchanged; killed by chloroform; autopsy showed everything normal.
- 3. Lieber (p. 14), where it is stated to be permitted by the Austrian law (pp. 22-23), where it is stated to be permitted by the Italian law (p. 31), where it is stated to be permitted by the French law in candies, pastilles, sweetmeats, sauces,

fruits, and certain liqueurs ordinarily not colored.

- 4. Weyl (p. 22), quoting Sonnenkalb (p. 24), quoting Grandhomme.
- 5. Fraenkel (p. 580) states that it is effective in only 5 per cent of malaria cases.
- 6. Permitted by the law of Austria.
- 7. Buss lists it as nonpoisonous.

### UNFAVORABLE.

1. Weyl (p. 23), quoting Friedrich, where poisoning was produced in a young man engaged in packing this dye.

## G. T. 459.

Trade names.—Iodin Green; Pomona Green; Night Green; Vert Lumière.

Scientific name.—Zinc-double-chlorid of heptamethyl-rosanilin-chlorid.

Discovered and patented.—1866.

Shade.—Green. Not offered.

FAVORABLE.

Nothing.

### UNFAVORABLE.

1. Chlopin (p. 175) on his own experiments classes it as "suspicious." (See p. 181). The experimental data are as follows:

Experimental data by Chlopin.

[1 gram=167 mg=117 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 9 10 11 12 13 14 15 16 17 18 Total.	Grams. 6 2 2 6	Kilos. 6	cc. 420 470 441 420 390 400 442 370	Before experiment dog and urine normal. Do. Urine slightly greenish; no albumen. Do. Urine has greenish opalescence; traces of albumen. Do. Urine has greenish opalescence; no albumen. Do. Urine has greenish opalescence; traces of albumen. Normal color; no albumen.

2. Buss lists it as poisonous.

### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

## G. T. 462.

Trade names.—Acid Magenta; Acid Fuchsin; Acid Rubin; Fuchsin S; Acid Rosein; Rubin S.

Names under which it was offered on the United States market as a food color in 1907.—Acid Magenta Powdered; Acid Magenta.

Scientific name.—Mixture of the sodium or ammonium salts of the trisulphonic acids of rosanilin and pararosanilin.

Discovered.—1877.

Shade.—Red. Offered by 2 out of 12 sources.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. CAZENEUVE (Arch. gén. méd., 1886, p. 753) says it may be taken without effect by man and animals, sick or well, in large doses.
- 3. Weyl (p. 55), where he says that it is established of this color that it is almost without poisonous action.
- 2. Lieber (p. 14), where it is stated to be permitted by the law of Austria (p. 31), where it is stated to be permitted by the law of France for confectionery, cordials, and the like.
- 5. Permitted by the law of Austria.
- 6. Permitted by the law of Italy.
- 7. CAZENEUVE AND LÉPINE (Compt. rend., 1885, v. 101, p. 1011): A. Dog: 15 kilos weight, received—

Days.	Grams.	Milligrams per kilo.	Grains per 100 pounds.
15	1	67	47
5	2	134	94
5	5	335	235
5	10	670	470

No diarrhea; no vomiting; no albuminuria; urine colored only occasionally, but did contain the leuco compound of the dye. B. Man: 1. Afflicted with Bright's disease; took two grams daily for one week; no effect. 2. Afflicted with renal cirrhosis; four grams daily for several days; no effect. 3. A well man took four grams daily for several days; no effect.

8. CAZENEUVE AND LÉPINE (Bull. de l'acad. de méd. 1886, p. 643): Tolerated by man, sick or well.

## G. T. 467.

Trade name.—Acid Violet 6B.

Scientific name.—Sodium salt of dimethyl diethyl dibenzyl triamido triphenyl carbinol disulphonic acid.

Discovered and patented.—1889.

Shade.—Violet. Not offered.

### FAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 54): A dog weighing 4,250 grams received 12.5 grams dye in 28 days, which amounts to 105 milligrams per kilo per day, or 74 grains per 100 pounds per day. The dog vomited the color only two or three times and was otherwise normal. Appetite, temperature, and urine all remained normal. Conclusion: Nonpoisonous; autopsy also showed everything normal.

UNFAVORABLE.

Nothing.

## G. T. 477.

Trade names.—Alkali Blue; Nicholson Blue; Fast Blue.

Scientific name.—Mixture of sodium salts of triphenyl rosanilin monosulphonic acid and of triphenyl para rosanilin-monosulphonic acid.

Discovered and patented.—1862. Shade.—Blue. Not offered.

### FAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 45): A dog weighing 4,500 grams received 25 grams dye in 30 days, which amounts to 185 milligrams per kilo per day, or 129.5 grains per 100 pounds per day. The urine remained of normal color, but the stool was a deep blue black. Throughout the whole time the animal was in perfect health. Killed with chloroform; autopsy showed everything normal.

UNFAVORABLE.

Nothing.

## G. T. 478.

Trade names.—Bavarian Blue DSF; Methyl Blue, water-soluble; Navy Blue B; Methyl Blue for silk MLB.

Scientific name.—Sodium salt of triphenyl pararosanilin di- and tri-sulpho acid.

Discovered.—1862; not patented.

Shade.—Blue. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 170) examined this color, and on his own experiments classed it as "nonpoisonous but not quite indifferent."

Experimental data by Chlopin.

[1 gram=115 mg=81 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 18 19 23 24 25 26 27 28 29 30 31 June 1 2 3 4 5 6 6 7 8 9 10 11-15 16 17-29 Total.	Grams. 2 2 2 2 2 3 3 3 3 3 3 3 2 2 2 2	Kilos. 8.7	cc.  450 365 380 370 370 320 525 327 405 395 353 370 340 1400 425	Dog and urine quite normal. Urine slightly greenish; no albumen; acid. Do. Do. Do. Do. Do. Do. Do. Do. Urine slightly greenish; traces of albumen; acid. Do. Do. Do. Do. Urine slightly greenish; traces of albumen; acid; diarrhea. Do. Urine slightly greenish; traces of albumen; acid; diarrhea. Do. Urine slightly greenish; traces of albumen; acid. Do. Do. Color normal; traces of albumen. Do. Do. Do.

## G. T. 479.

Trade names.—Methyl Blue O; Brilliant Cotton Blue greenish; XL Soluble Blue; Diphenylamin Blue; Bavarian Blue DBF; Soluble Blue 8B and 10B; Helvetia Blue.

Scientific name.—Sodium salt of triphenyl-pararosanilin trisulphonic acid.

Discovered and patented.—1862. Shade.—Blue. Not offered.

21000 1100 0=====

Nothing.

FAVORABLE.

UNFAVORABLE.

1. Chlopin (p. 168) examined this color and on his own experiments classed it as "suspicious" or "nonsuspicious" dependent upon the make of goods.

Experimental data by Chlopin.

No. 1.

[1 gram=75 mg=52 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 14 15	Grams.	Kilos. 13.4	cc. 460	Dog normal; no albumen. Color chocolate-brown; insignificant traces of albumen.
16 17	2		410	Color same; no albumen. Urine normal; no albumen; dog is well.
Total	4			

No. 2.

[1 gram=152 mg.=106 grains.]

1901. Nov. 12 13	3	6.6	300	Dog quite well; urine normal color; acid; no albumen.
15 16 17 19 20	3 3 3 3		360 305 330	Do. Do. Urine scarcely perceptible greenish sheen; acid; no albumen. Urine greenish color; no albumen. Urine darker; no albumen.
21 22 Total	15	6.8		Urine color normal; no albumen. Color almost normal.

This preparation from Moscow; not suspicious.

No. 3 (MERCK'S PREPARATION).

[1 gram=147 mg=103 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1903.				
Feb. 18	3	6.9	400	Before experiment dog quite normal; urine acid; no albumen; normal yellow color; 2 hours after giving dye vomiting.
19	3			Vomiting stopped; diarrhea; urine chocolate-brown; reaction acid; considerable albumen; appetite not decreased.
20	3		330	During night diarrhea; no vomiting; urine brown; acid; traces of
21	3			albumen; without acid urine becomes blue.  No diarrhea; no vomiting; urine brown; traces of albumen.
22 23	3	6.8		Urine brown, with blue sheen; traces of albumen. Urine blue chocolate-brown; otherwise normal. Urine acquired
Total.	17			usual color 8 days later.
Total	15			

Conclusion: "Suspicious."

## G. T. 480.

Trade names.—Soluble Blue; China Blue; Cotton Blue; Bleu Marine; Water Blue; Water Blue 6 B extra; London Blue extra.

Names under which it was offered on the United States market as a food color in 1907.—Pure Soluble Blue.

Scientific name.—Sodium, ammonium or calcium salt of the trisulphonic acid (with some disulphonic acid) of triphenyl-rosanilin and triphenyl-pararosanilin.

Discovered and patented.—1862.

Shade.—Blue. Offered by 1 out of 12 sources.

### FAVORABLE.

1. Permitted by law of Austria.

- 2. LIEBER (p. 147): A guinea pig received 306 milligrams per kilogram body weight, or 214 grains per 100 pounds, once a day seven times every other day; the weight remained substantially constant, a slight gain of less than \(\frac{3}{4}\) per cent being noted. "Nothing irregular or disturbing whatsoever was observed during the whole period."
- 3. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 45): A dog weighing 4,500 grams received 30 grams dye in 30 days which amounts to 223 milligrams per kilo per day or 156 grains per 100 pounds per day. Animal remained well throughout; no loss of weight or appetite; urine normal color, stool deep blue; killed with chloroform; autopsy showed a pea-green coloring of the cortex of the kidneys.

#### UNFAVORABLE.

 FRAENKEL (p. 574), quoting Penzoldt, says that it completely arrests germ development, and causes changes internally.

2. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 46): Regards this as injurious hypodermically, but not through mouth. A dog weighing 4,000 grams received 3.5 grams of dye hypodermically in 16 days, at the end of which time he died; this amounts to 55 milligrams per kilo per day or 38.5 grains per 100 pounds per day. The autopsy showed the liver to be free from blood; kidneys soft and congested; all organs swollen and colored.

## G. T. 483.

Trade names.—Aurin; Rosolic Acid; Yellow Corallin.

Scientific name.—Mixture of aurin (trioxytriphenyl-carbinol) oxidized aurin, methylaurin, and pseudo-rosolic acid (corallin phthalin).

Discovered.—1834.

Shade.—Yellowish brown. Not offered.

### FAVORABLE.

1. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): Rosolic Acid is positively non-poisonous.

#### UNFAVORABLE.

1. Chlopin (p. 167) examined this color, and on his own experiments classes it as "strongly poisonous." The experimental data are as follows:

### [1 gram=137 mg=96 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 12 13 14 15 16 Total	Grams. 1 2 3 3 3	Kilos. 7.3	cc. 330 375 300	Before experiment dog quite well; urine normal color; acid; no albumen. Diarrhea; urine faint red. Diarrhea; lassitude; eats little; urine red; no albumen. Violent diarrhea and vomiting; dog stands on feet with difficulty; eats nothing. Same conditions as on preceding day; dog sick for a long time after.

- 2. Prohibited by the German law of 1887.
- 3. Prohibited by the Belgian law of June 17, 1891.
- 4. Buss lists it as poisonous.

### DOUBTFUL.

1. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): Rosolic Acid is positively nonpoisonous. Small animals can take 1 gram and more of it. In Austria it is
prohibited for use in coloring foods. Corallins, or red (Pæonin) or yellow
colors, consisting of Aurin and Rosolic Acid were regarded as poisonous
because in experiments on man and animals illness occurred, but are said to
be poisonous only in the presence of arsenic, phenol, or anilin. These sub
stances are prohibited in the coloring of food.

## G. T. 488 or 490.

Trade name.—Victoria Blue; Victoria Blue B; Victoria Blue 4R. Scientific names.—Hydrochlorid of phenyltetra (penta) methyltriamido-diphenyl-alpha-naphthyl-carbinol (note: 4R is bracketed).

Discovered and patented.—1883.

Shade.—Blue. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 47): A dog weighing 5,250 grams received 10.5 grams dye in 22 days, which amounts to 45 milligrams per kilo per day or 32 grains per 100 pounds per day. After receiving 1.5 grams the animal suffered copious continuous salivation, anemia, and emaciation and occasional vomiting. The urine's color did not change; the feces became blue; temperature slightly above and below normal; although highly emaciated (loss in weight was 1,750 grams or 33½ per cent) the animal retained its appetite; animal died on the morning of the 23d day. The autopsy showed œsophagus, stomach, and intestines colored deep blue and filled with a greenish scum, extended and strong catarrh of the stomach and intestines; kidneys and liver contained very little blood. Conclusion: Poisonous.

(Note: It is uncertain which of these two dyes Santori used.)

## G. T. 502.

Trade name.—Rhodamin G and G extra.

Scientific name.—Chiefly Triethylrhodamin.

Discovered and patented.—1891.

Shade.—Bluish. Offered by 2 out of 12 sources.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 184) examined this color, and on his own experiments classes it as "suspicious." The experimental data are as follows:

Experimental data by Chlopin.

[1 gram=167 mg=117 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1903. Feb. 18	Grams.	Kilos.	cc. 310	Dog normal; urine acid; no albumen; after 2 hours thin stool; remainder of day lively; good appetite. Urine fuchsin color, acid, no albumen; stool and appetite normal. Goes to stool without results; otherwise as above.
21 22 23	3 3		270 320	Do. Do. Goes to stool without results; urine normal after 8 days.
Total	15			

## G. T. 504.

Trade names.—Rhodamin B; Rhodamin O; Safranilin.

Names under which it was offered on the United States market as a food color in 1907.—Rhodamin B extra; Rhodamin; Rhodamin B.

Scientific name.—Hydrochlorid of diethylmeta-amido-phenol-phthalein.

Discovered and patented.—1887.

Shade.—Bluish red. Offered by 5 out of 12 sources.

### FAVORABLE.

1. Lieber (p. 141): A young female rabbit received 339 milligram body weight, or 237 grains per 100 pounds, five times on alternate days. "During the whole period the animal seemed to be perfectly at ease, was lively, displayed good appetite, and gained steadily \* \* \* ." The gain in weight was, roughly, 7 per cent.

### UNFAVORABLE.

1. Chlopin (pp. 182, 183) examined this color, and on his own experiments classifies it as "not poisonous, but not entirely indifferent; suspicious." The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram = 109 mg = 76 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1904. Feb. 18 19 20 21 22 Total.	Grams. 3 3 3 3 3 3 15	Kilos. 9. 2	420 400 400 420	Dog normal; urine acid, no albumen; strong yellow color; 2 hours after giving color thin stool. Urine colored wine red, strong fluorescence, disappears on boiling, and reappears on cooling; no albumen. Thin stool; no change in other respects. No diarrhea; no change in other respects. General condition good; urine red and acid; no albumen.

Dog recovers.

## G. T. 512.

Trade names.—Eosin; Eosin A; Eosin B; Eosin A extra; Eosin Yellowish; Eosin G G F; Water-soluble Eosin; Eosin 3 J and 4 J extra.

Names under which it was offered on the United States market as a food color in 1907.—Erythrosin I N; Eosin J; Eosin Y.

Scientific name.—Alkali salts of Tetra-bromo-fluorescein.

Discovered.—1874.

Shade.—Yellowish red. Offered by 3 out of 12 sources.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 31): "According to Grandhomme, rabbits bear without injury \* \* \* Eosin \* \* \*."
- 3. Permitted by the law of Austria.
- 4. Buss lists it as nonpoisonous.

#### UNFAVORABLE.

- 1. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): "The continued use of these coloring matters, as well as of Phenolphthalein, which becomes colored in the system, I regard as harmful, and, in fact, through action as coloring matters."
- 2. Forbidden by the Italian law.

## G. T. 516.

Trade names.—Erythrosin G; Pyrosin J; Jaune d'Orient; Dianthin G; Iodeosin G.

Name under which it was offered on the United States market as a food color in 1907.—Erythrosin yellowish shade.

Scientific name.—Sodium or potassium salt of diiodofluorescein.

97291°-Bull. 147-12-9

Discovered.—1875.
Shade.—Yellowish red. Offered by 1 out of 12 sources.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 181): Examined this color, and on his own experiments reports it as "injurious because of light albuminuria, vomiting, and diarrhea." The experimental data are as follows:

Experimental data by Chlopin.

No. 1.

[1 gram = 156 mg = 109 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 24 25 26 27 28 29 30 May 3	Grams. 2 2 4	Kilos. 6. 4	cc. 360 353 368 370 355 365 365	Before experiment dog and urine normal. Urine red yellow with greenish sheen; acid; no albumen. Urine red yellow with greenish sheen; acid; trace of albumen. Do. Urine red yellow with greenish sheen; acid; no albumen. Do. Do. Do. Do.

No. 2.

### [1 gram = 152 mg = 106 grains.]

1901. Nov. 24 25 26 27 28	3 3 3 3	6.6	Do. Urine fluorescent orange; r Vomiting, diarrhea; urine 380 No vomiting and no diarr	
29	3		340	Do.
30				Do.
Dec. 2-3				Color and composition of urine normal; dog well.
Total.	15			

### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm. 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 517.

Trade names.—Erythrosin; Erythrosin B; Iodeosin B; Eosin J; Erythrosin D; Pyrosin B; Eosin Bluish.

Names under which it was offered on the United States market as a food color in 1907.—Erythrosin Yellow Shade; Erythrosin B; Erythrosin.

Scientific name.—Sodium or potassium salt of tetraiodofluorescein. Discovered.—1876.

Shade.—Bluish red. Offered by 5 out of 12 sources.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Weyl (p. 31): "According to Grandhomme, rabbits bear without injury \* \* \* Erythrosine \* \* \*."
- 3. Permitted by the laws of France.
- 4. Permitted by the law of Austria.
- FRAENKEL (p. 574): "Rose Bengal \* \* \* produces no noticeable disturbances."
- 6. Buss lists it as nonpoisonous.

#### UNFAVORABLE.

1. Lewin (Lehrbuch der Toxikologie, 1897, p. 231): "The continued use of these coloring matters, as well as of phenolphthalein, which becomes colored in the system, I regard as harmful, and, in fact, through action as coloring matters."

### G. T. 520.

Trade names.—Rose Bengal; Rose Bengal A T; Rose Bengal N; Rose Bengal G.

Names under which it was offered on the United States market as a food color in 1907.—Rose Bengal B; Phloxin B.

Scientific name.—Alkaline salt of tetraiododichlorofluorescein.

Discovered.—1875.

Shade.—Bluish red. Offered by 2 out of 12 sources.

### FAVORABLE.

1. Permitted by Confectioners' List.

### G. T. 521.

Trade names.—Phloxin; Phloxin TA; Eosin 10B.

Scientific name.—Sodium salt of tetrabromotetrachlorofluorescein. Discovered.—1882.

Shade.—Red. Not offered.

### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Permitted by the Austrian law.
- 3. Chlopin (p. 185) examined this color, and on his own experiments classifies it as "nonpoisonous." The experimental data are as follows:

### Experimental data by Chlopin.

No. 1. [1 gram=143 mg=100 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 10 11 12 13 14 15	Grams. 1 3	Kilos. 7. 0	cc. 380 370 370 370 370	Dog and urine quite normal.  Do.  Do.  Do.  Urine orange.  Do.

No. 2.

### [1 gram=116 mg=81 grains.]

1901, Nov. 3 4 5 6 7 8 9 10 11 12-14	3 3 3 3 3	8.6	450 430 400 440 430 450 400	Dog well; urine normal color; acid; no albumen. Urine orange; no albumen. Do. Do. Do. Do. Do. Do. Color almost normal; no albumen. Do.	
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## G. T. 527.

Trade names.—Cœrulein S; Alizarin Green; Anthracene Green. Scientific name.—Sodium bisulphite compound of cœrulein. Discovered.—1879.

Shade.—Black. Not offered.

### FAVORABLE.

1. Chlopin (pp. 186-7) examined this color, and on his own experiments reports it as "nonpoisonous, and not sufficient data to regard it as suspicious." The experimental data are as follows:

Experimental data by Chlopin.

No. 1. [1 gram=147 mg=103 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Arug. 30 31 Sept. 1 2 3 4 5 6 7 9 10 11 12-18	Grams. 2 3 3 3 3 3 3 3 2 2 22	Kilos. 6.8	cc. 320 330 300 330 300 310 300 375	Before experiment dog quite normal. Urine normal; acid; no albumen. Do. Urine has scarcely perceptible sheen; no albumen. Do. Do. Do. Do. Do. Nothing abnormal. Do. Do. Do. Do. Do. Do. Do. Do.

## Experimental data by Chlopin—Continued.

### No. 2.

### [1 gram=167 mg=117 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 3 4 5 6 Total	Grams. 2 3 3 2 2 10	Kilos.	cc. 270 250 265	Before experiment dog quite well; urine normal. Urine yellow with orange sheen; acid; no albumen. Thin stool twice; urine dark yellow with greenish sheen; acid; no albumen. Stool normal; urine yellow green; acid; no albumen; general condition quite well.

### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 530.

Trade name.—Benzoflavin.

Scientific name.—Hydrochlorid of diamido-phenyl-dimethyl-acridin. Discovered.—1887.

Shade.—Brownish orange yellow. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 189) on his own experiments regards this color as suspicious. The experimental data are as follows:

### Experimental data by Chlopin.

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. May 3 4 5 6 7 8 9	Grams. 2 2 2 6	Kilos.	cc. 365 391 330 375	Before experiment dog well and urine normal. Dog ate poorly; traces of albumen. Do. Urine greenish sheen; acid. Color same; no albumen. Do. Do.

### G. T. 532.

Trade names.—Phosphin; Xanthin; Leather Yellow; Philadelphia Yellow G.

Scientific name.—Nitrate of chrysanilin (unsym. diamido-phenylacridin) and homologues.

Discovered.—1862.

Shade.—Orange yellow. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 190) reports Fuchsin containing Phosphin as suspicious.

2. On his own experiments says this color "does not belong to the poisonous colors, but is not wholly harmless." The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram=147 mg=103 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Apr. 15 16 17 18 19 21 22 23 24 Total	3 2 2 7	Kilos. 6.8	cc. 390 350 350 350 330 370	Dog well; urine normal.  Do.  Do.  Urine yellower than normal; vomited several times after taking dye; no albumen.  Urine same; no vomiting.  Vomited twice; urine same.  No vomiting; urine same.  Color normal.  Dog well; urine normal.

3. (p. 178): See experimental data on G. T. 448, which also applies to this color.

4. Fraenkel (p. 578): Where its physiological action is compared with quinin its action on protozoa is far greater. "The Phosphins are locally strong irritants, and producers of inflammation of medium poisonous nature so that humans can very well bear 400 milligrams, or 6.17 grains.

5. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Phosphin \* \* \* produces

in humans, in doses up to 1 gram, vomiting and diarrhea."

### DOUBTFUL.

 WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

## G. T. 563.

Trade names.—Alizarin Blue S; Anthracene Blue S; Alizarin Blue ABS.

Scientific name.—Sodium bisulphite compound of dioxyanthra-quinone-beta-quinolin.

Discovered and patented.—1881.

Shade.—Blue. Not offered.

### FAVORABLE.

1. Permitted by law of Austria.

### UNFAVORABLE.

1. Chlopin (p. 171): On authority not given reports this color as poisonous or harmful.

2. Ehrlich (Das Sauerstoff beduerfniss des Organismus, 1885, p. 23): "Per kilogram of rabbit, 12-15 cc of this solution in general produce death within the first quarter of an hour; whereas 4 cc of the same did not usually produce it, and 7 cc represent a medium, when properly applied, fatal dose." (This solution contained not to exceed 17 per cent coloring matter; each cubic centimeter represents 170 milligrams per kilogram body weight, or 119 grains per 100 pounds; the coloring matter was introduced subcutaneously.)

3. Buss lists it as poisonous.

## G. T. 572.

Trade name.—Indophenol white; Leucindophenol.

Scientific name.—Tin compound of dimethyl-para-amido-phenyl-para-oxy-alphanaphthylamin.

Discovered.—1881.

Shade.—Blue. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 57): A dog weighing 4,000 grams received 18 grams dye in 30 days, which amounts to 150 milligrams per kilo per day or 105 grains per 100 pounds per day. Temperature, urine, and weight all remained unchanged. Animal killed with chloroform; autopsy showed fatty degeneration of the liver; everything else normal. (Santori classes this color as "not nonpoisonous.")

## G. T. 574.

Trade names.—Ursol D; Ursol P; Ursol DD.

Scientific name.—Hydrochlorids of para-phenylene diamin, para-amidophenol, and diamidodiphenylamin, respectively.

Discovered.—1888.

Shade.—Brown to black. Not offered.

FAVORABLE.

Nothing.

### UNFAVORABLE.

1. Chlopin (p. 214) examined this color, and on his own experiments reports it as "strongly poisonous." On January 15, 1901, a dog weighing 18.4 kilos was given 3 grams. An hour or an hour and a half after giving dye vomiting set in; dog lay down on one side and died in 3 or 4 hours. Cause of death, heart filled with coagulated blood; lungs, liver, and kidneys filled with blood; turbid swelling of the liver and heart; mucous membrane of stomach inflamed; brain unchanged. Death caused by paralysis of the heart. It also acts severely on the skin.

### G. T. 576.

Trade names.—New Gray; Malta Gray; Nigrosin; Direct Gray; Methylene Gray; New Methylene Gray.

Scientific name.—(?)

Discovered.—1888.

Shade.—Gray. Not offered.

### FAVORABLE.

1. Chlopin (p. 209) examined this color, and on his own experiments concludes it contains "no poisonous properties." The experimental data follow:

### Experimental data by Chlopin.

[1 gram=77 mg=54 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Feb. 8 9 10 11 12 13 14 15 16 17	Grams. 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3	Kilos. 12.9	cc. 510 480 400 450 475 490	Dog quite well; urine quite normal. Urine green color; acid; no albumen. Do. Do. Vomited once; urine dark green; acid; no albumen. Do. Do. Urine lighter shade. Urine normal; dog well.

## G. T. 584.

Trade names.—Safranin; Safranin S; Safranin conc.; Safranin GOO; Safranin T; Safranin extra G; Safranin FF extra; Safranin AG, AGT, and OOF.

Name under which it was offered on the United States market as a food color in 1907.—Safranin SP.

Scientific name.—Mixture of diamido-phenyl-and-tolyl-tolazonium chlorids.

Discovered and patented.—1859.

Shade.—Reddish brown. Offered by 1 out of 12 sources.

### FAVORABLE.

1. CAZENEUVE (Arch. gén. de méd. 1886, Vol. I, p. 753): Produces gastro-intestinal disturbances but is not a violent poison.

#### UNFAVORABLE.

- 1. Prohibited by Confectioners' List.
- Weyl (p. 31): "Cazeneuve and Lépine pointed out the poisonous nature of \* \* \* Safranine."
- 3. "This body (Chamber of Commerce of Sonneberg) recommends for the preparation of children's toys three colors, the poisonous character of which I can demonstrate. These are \* \* \* Safranine \* \* \* . (p. 34.)
- 4. Weyl (Handbuch der Hygiene): "According to Theodore Weyl this is even in small doses, when injected subcutaneously, a strong poison" (50 milligrams per kilo body weight, or 35 grains per 100 pounds); "whereas, when administered by the stomach only large doses over a long period of time produce diarrhea."
- 5. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Safranin is poisonous when injected intravenously. (Pulse acceleration, dyspnœa, cramps.) Fed to dogs by the mouth it causes only diarrhea."
- Prohibited by the Resolutions of the Society of Swiss Analytical Chemists, of September, 1891.
- 7. Prohibited by the Canton of Tessin.
- 8. Buss lists it as poisonous.
- 9. CAZENEUVE AND LÉPINE (Compt. rend. 1885, v. 101, p. 1011): A dog (weight not given) was given, by the mouth, daily doses of from 1 to 4 grams for several weeks; only salivation and diarrhea were produced. They conclude that it is a harmful color.

#### DOUBTEUL.

1. WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it almost completely inhibits digestion.

# G. T. 593.

Trade names.—Mauve; Mauvein; Chrome Violet.

Obsolete names.—Mauve Dye; Anilein; Anilin Purple; Violein; Indisin.

Scientific name.—Salts of phenyl- and tolyl-safranins.

Discovered and patented.—1856.

Shade.—Blue, Reddish Blue, Bluish Violet. Not offered.

#### FAVORABLE.

1. Permitted by Confectioners' List.

# G. T. 599.

Trade names.—Indulin, spirit soluble; Indulin opal; Fast Blue R and B, spirit soluble; Indulin 3B and 6B, spirit soluble; Indulin 3B opal; Indulin 6B opal; Azin Blue, spirit soluble; Indigen D and F; Printing Blue; Acetin Blue.

Scientific name.—Mixtures of dianilidoamido, trianilido and tetraanilido phenyl-phenazonium chlorids.

Discovered and patented.—1863.

Shade.—Blue. Not offered.

#### FAVORABLE.

Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 50): A dog weighing 4,600 grams received 12.5 grams of this dye (in oil) in 30 days, which amounts to 91 milligrams per kilo per day, or 64 grains per 100 pounds per day. There was no disturbance of any kind. Killed by chloroform; autopsy showed everything to be normal.

# G. T. 600.

Trade names.—Nigrosin, spirit soluble; Coupier's Blue; Oil Black; Sloelin; Spirit Black.

Scientific name.—Mixtures of Indulins with allied bases and fluorindins.

Discovered and patented.—1867.

Shade.—Black. Not offered.

#### FAVORABLE.

1. Permitted by Confectioners' List:

# G. T. 601.

Trade names.—Indulin, soluble; Indulin 3B; Fast Blue R and 3R; Sloelin R S and B S; Indulin R and B; Indulin 6 B; Fast Blue 2R, B, and 6B.

Scientific name.—Mixtures of the sodium salts of the sulphonic acids of the various spirit-soluble indulins.

Discovered and patented.—1867.

Shade.—Bronzy or Blue Black. Offered by 1 out of 12 sources.

#### FAVORABLE.

1. Chlopin (pp. 198-9): On his own experiments classifies this color as nonpoisonous. The experimental data are as follows:

Experimental data by Chlopin.

No. 1. [1 gram=54 mg=38 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.	
1902. Jan. 5 6 7 8 9 10 11 12 13 14 Total.	Grams. 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3 3	Kilos. 18. 4	cc. 650 670 400 410 400 400 500 560 650 620	Dog and urine normal. Urine brown; acid; no albumen. Urine greenish brown; no albumen. Do. Do. Do. Do. Do. Do. Do. Color and composition of urine normal; dog is well.	

No. 2.  $[1 \ \mathrm{gram}{=}114 \ \mathrm{mg}{=}80 \ \mathrm{grains.}]$ 

1902. Jan. 14 15 16 18 19 20 21 22 23 24 25 26	3 3 3 3 3 3 3 3	8.8	420 390 420 400 460 400 450 400	Dog and urine normal. Urine slightly blue; acid; no albumen. Do. Do. Urine almost normal color; no albumen. Color and other properties of urine normal. No record. Urine slightly bluish; no albumen. Do. Dark green; no albumen. Feeble green; no albumen. Urine normal; dog well.
Total.	18			

#### UNFAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 55): A dog weighing 4,500 grams received 4 to 6 grams dye in 7 days, which amounts to 127 to 190 milligrams per kilo per day, or 89 to 133 grains per 100 pounds per day. No vomiting. Stool black blue and no change in the urine. Up to the sixth day the animal, in very good general condition, ate heartily and was quite lively; temperature unchanged. On the morning of the sixth day the animal was found in his cage suffering from general muscular cramps which were heightened by the slightest noise; the animal did not respond to calls or threatening movements and was in a complete stupor. This continued for 24 hours, when the animal died. The autopsy showed numerous punctures of the lungs and of the mucous membrane of the stomach; fatty degeneration of the liver and little blood in it; kidneys without change, although the cortex was colored a light green.

# G. T. 602.

Trade names.—Nigrosin, soluble; Bengal Blue; Gray R and B. Scientific name.—Sodium salts of sulphonic acids of spirit nigrosins.

 $Discovered\ and\ patented. {\color{red}\longleftarrow} 1867.$ 

Shade.—Black. Not offered.

FAVORABLE.

Nothing.

UNFAVORABLE.

1. Lewin (Lehrbuch der Toxikologie, 1897, p. 231) says: "Produces eczema," and cites Deutsche. Med. Wochenschr., 1891, page 45.

# G. T. 614.

Trade names.—Magdala Red; Naphthalene Rose; Sudan Red; Naphthalene Red; Naphthylamin Pink.

Scientific name.—Mixture of amido-naphthyl-naphthazonium-chlorid and diamido-naphthyl-naphthazonium chlorid.

Discovered.—1868.

Shade.—Red. Not offered.

FAVORABLE.

Nothing.

UNFAVORABLE.

 CHLOPIN (p. 200) examined this color, and on his own experiments concludes it "does not belong to the poisonous list, but is not wholly harmless." The experimental data are as follows:

Experimental data by Chlopin.

No. 1.  $[1~{\rm gram}{=}91~{\rm mg}{=}64~{\rm grains.}]$ 

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Apr. 12 13 14 15 18	Grams. 2	Kilos.	780 1,100 760 660 670	Before experiment dog and urine normal. Urine rose-colored; acid; no albumen. Do. Urine quite red; acid; no albumen; vomited once. No vomiting; dog is well; urine normal in color, and composition.
Total.	5			

No. 2. [1 gram=111 mg=78 grains.]

1903. Feb. 3 4 5 66 7 8 9	3 3 3 3	9	380 340 370 310 320	Day before experiment quite well; urine normal color; acid; no albumen.  In the morning three times thin stool, urine faintly rose-colored; acid; no albumen.  No diarrhea; urine rose-red; acid; no albumen.  Do.  Do.  Do.  Do.  Do.  Do.
Total .	15			

# Experimental data by Chlopin—Continued.

[1 gram=114 mg=80 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1903. Feb. 10	Grams.	Kilos.	cc. 310 315 350	Vomiting and diarrhea; urine rose-red; no albumen; acid; with muriatic acid rose color, becomes greenish gray.  No vomiting; no diarrhea; urine rose-red; acid; no albumen.  No vomiting; no diarrhea; color reaction of urine normal; no albumen.
Total.	3			

#### DOUBTFUL.

1. Winograpow (Zts. Nahr. Genussm. 1903, v. 6, p. 589) says it almost completely inhibits digestion.

# G. T. 620.

Trade names.—Gallocyanin DH and BS; Fast violet; Gallocyanin RS, BS, and D.

Scientific name.—Dimethylamido dioxy phenazoxoniumcarboxylate. (BS is the bisulphite compound.)

Discovered and patented.—1881.

Shade.—Bluish Violet. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 51): A dog weighing 5,400 grams received 7.5 grams dye in 30 days, which amounts to 46 milligrams per kilo per day or 32 grains per 100 pounds per day. Throughout all the time the animal remained well, had good appetite, temperature normal, no loss of weight; urine and feces colored deep blue black. Killed by chloroform. Autopsy showed incipient fatty degeneration of the liver and a swelling of the kidneys. Conclusion: Poisonous.

# G. T. 639.

Trade names.—Meldola's Blue; Cotton Blue R; Fast Navy Blue R; Naphthol Blue R and D; Naphthylene Blue R in crystals; Fast Blue R, 2 R and 3 R for cotton in crystals; Fast Navy Blue RM and MM.

Scientific name.—Zinc double chlorid of dimethylamido-naphthophenoxazonium chlorid.

Discovered.—1879.

Shade.—Dark Violet. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (pp. 194-195) on his own experiments reports this color as "very poisonous." The experimental data are as follows:

# Experimental data by Chlopin.

## No. 1.

## [1 gram=68 mg=48 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Jan. 5	Grams.	Kilos. 14. 6	cc. 590	Before experiment dog quite well; urine normal in color and composition; 1 hour after giving the dye vomiting began; dog lay down on ground and died in 1 hour.
Total.	3			

#### No. 2.

### [1 gram=167 mg=117 grains.]

1902. Jan. 14	3	6	 Soon after giving dye, strong vomiting and diarrhea; after a few hours improved.  Most violent vomiting; dog lay down on his side, and was found dead in 6 hours.
Total.	6		

Note.-See p. 181.

2. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 49): A dog weighing 4,506 grams received 12.5 grams dye during 30 days; this amounts to 93.3 milligrams per kilo per day or 65.3 grains per hundred pounds per day. Continued vomiting beginning with 0.2 grams dye, anaemia and copious as well as continuous salivation and emaciation; the animal lost 1,200 grams in weight or 27 per cent. Killed by chloroform; autopsy showed no fat, flabby muscles, stomach contracted and filled with mucous and in part colored pea green; fatty degeneration of the liver; contracted bladder; kidneys swollen and congested with blood and decomposed blood corpuscles in the Bowman capsules.

# G. T. 649.

Trade names.—Gentianin; Gentiana Violet.

Scientific name.—Zinc double chlorids of dimethyldiamido phenazthionium chlorid.

Discovered and patented.—1886.

Shade.—Violet. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 54): A dog weighing 3,006 grams received 4.7 grams of dye in 7 days, which amounts to 224 milligrams per kilo per day or 157 grains per 100 pounds per day. Beginning with the third day the dog appeared weak and depressed and a whitish froth appeared at the mouth; mild diarrhea and complete aversion to food; temperature and urine unchanged. Died on seventh day. Autopsy showed congestion of mucous membrane of stomach; the liver was inflamed and the kidneys strongly congested.

# G. T. 650.

Trade names.—Methylene Blue B and BG; Methylene Blue BB in powder extra D; Methylene Blue BB in powder extra; Methylene Blue A extra.

Names under which it was offered on the United States market as a food color in 1907.—Methylene Blue B; Methylene Blue.

Scientific name.—Chlorid or zinc double-chlorid of tetramethyldiamido-phenazthionium.

Discovered.—1876.

Shade.—Blue. Offered by 2 out of 12 sources.

#### FAVORABLE.

- 1. Schacherl (p. 1046): "To these groups belong the much-used Methylene Blue, which in moderate doses is harmless."
- 2. Fraenkel (p. 574): "\* \* Methylene Blue causes no noteworthy disturbances."
- 3. CAZENEUVE (Arch. gén. de. méd. 1886, v. 1, p. 753) says that it produces gastric intestinal derangements but is not a violent poison.

#### UNFAVORABLE.

- 1. Prohibited by Confectioners' List.
- 2. Weyl (p. 31): "Cazeneuve and Lépine pointed out the poisonous nature of \* \* \* Methylene Blue \* \* \*."
- 3. Fraenkel (p. 579): "To regard Methylene Blue as a specific remedy such as quinin, is, in spite of a few such experiments, improper; it produces subsidiary effects which depend in part upon local irritation of the intestinal tract, and partly, however, upon irritation of the bladder with increased micturition."
- 4. CAZENEUVE (Arch. gén. de méd. 1886, v. 1, p. 753), says that it produces gastric intestinal derangements.
- 5. Combemale and Francois (Sem. Med. 1890, no. 31, p. 258), say that it produces intestinal disorders and vomiting, colored urine, and colored feces in dogs, and therefore is a highly injurious color.
- 6. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 42) classes it as injurious. A dog weighing 4,600 grams received in 20 days 18 grams dye, which amounts to 196 milligrams per kilo per day, or 137 grains per 100 pounds per day. Urine and feces colored; diarrhea and continuous vomiting; blood and pus contained in stool; loss of appetite; loss of weight was 1,600 grams, or 35 per cent. Animal died. Autopsy disclosed blue-colored skin and fat; brain turned blue on exposure to air, but only the outer cortex was colored; stomachical catarrh; the mucous membrane of the stomach colored blue; the heart sac and the pleura colored blue to blue-green; intestines externally blue; kidneys were thickened and colored dark blue throughout; parenchymatous kidney inflammation; fatty degeneration of the liver; diaphragm locally colored.
- Arloing and Cazeneuve, quoting Cazeneuve and Lépine (Arch. de méd. v. 9, p. 364), say that it is not inactive.
- 8. Forbidden by Resolutions of Swiss Analytical Chemists, September, 1891.

- 9. Galliard (Rev. intern. des falsifications; abst. Hygien. Runsdach. 1892, p. 104):
  "Methylene Blue, which is frequently used for coloring foods, can cause, even in small doses (10 to 20 milligrams, or one-sixth to one-third of a grain), a feeling of general depression, nausea, and certain feelings of pain, and can even produce transitory albuminuria. In larger doses (40 to 60 milligrams, or sixtenths to nine-tenths of a grain), it causes in the case of persons not accustomed thereto, vomiting, diarrhea, increased micturition, and albuminuria. In the case of persons suffering from nervous diseases, it frequently produces disturbance which has as its consequence a cessation or a change of place of the pain. Sometimes it produces painlessness, or an easing of pain in the patient, for which no certain therapeutic indication could be determined."
- 10. Weyl (Handbuch der Hygiene) comments as follows: "These statements of Galliard arouse but little confidence, because, as is well known, Methylene Blue is very frequently administered to invalids in doses of more than 0.5 gram without any noticeable disturbance. Perhaps Galliard's preparation was unclean. At any rate, in all experiments on the poisonous nature of Methylene Blue it is to be considered that it frequently occurs in commerce as a zinc chlorid double salt."
- 11. Lewin (Lehrbuch der Toxikologie, 1897, p. 232): "Methylene Blue \* \* \* can produce after prolonged administration of 0.5-1.5 gram daily increased micturition, irritation of the bladder, blue coloration of the urine, and saliva, diarrhea, headaches, vertigo, delirium, and twitching of the muscles, the latter symptoms probably because the coloring matter is deposited in the brain."

12. Buss lists it as poisonous.

# G. T. 651.

Trade name.—Methylene Green G conc. extra yellow shade. Scientific name.—Nitromethylene Blue. Discovered and patented.—1886.
Shade.—Green. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (pp. 192-193) examined this color, and on his own experiments concludes that it is "nonpoisonous, but somewhat suspicious." The experimental data are as follows:

Experimental data by Chlopin.

No. 1. [1 gram=71 mg=50 grains.]

Date. I	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
Apr. 24 25	Frams.	Kilos.	cc. 580	Dog well; urine normal. Urine dark green; acid; no albumen.
26 28 29 30	2 .		407	Urine less colored; acid; traces of albumen. No record. Urine dark brown; no albumen; acid.
May 2	4		485	Do. Color normal; acid; no albumen.

# Experimental data by Chlopin-Continued.

#### No. 2.

## [1 gram=122 mg=85 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1901. Nov. 25 26 27 28 29 Dec. 1-2	Grams. 3 3 3 3 3 3 3 15	Kilos. 8.2	cc. 430 445 450 410 440	Dog well; urine normal. Urine strongly green; no albumen. Do. Diarrhea; urine green; acid; no albumen. No diarrhea; urine same. Color normal; acid; no albumen; dog lively and well.

#### DOUBTFUL.

1. WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p, 589), says it noticeably retards digestive action; is not indifferent.

# G. T. 654.

Trade name.—Toluidin Blue O.

Scientific name.—Zinc-double-chlorid of dimethyl-diamido-toluphenazthonium-chlorid.

Discovered and patented.—1888.

Shade.—Blue. Not offered.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

I. Fraenkel (p. 574): "Toluidin Blue \* \* \* is a strong poison for microorganisms, and may be used in eye treatment, like Methylene Blue."

# G. T. 659.

Trade names.—Primulin; Thiochromogen; Sulphin; Polychromin; Aureolin.

Scientific name.—Sodium salt of the mono-sulphonic acids of the dehydrothionated condensation products of dehydrothiotoluidin (mixed with some sodium dehydrothiotoluidin-sulphonate).

Discovered.—1887.

Shade.—Yellow. Not offered.

FAVORABLE.

Nothing.

# UNFAVORABLE.

1. Chlopin (p. 203) examined this color, and on his own experiments reports it as "suspicious." The experimental data are as follows:

## Experimental data by Chlopin.

[1 gram=154 mg=108 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 21 22 23 24 25 26	Grams. 2.0 3.0 10.5	Kilos. 6. 5	cc. 292 294 355 260	Dog and urine normal. Urine dark brown: insignificant traces of albumen; acid. Color normal; no albumen. Urine dark brown; acid; no albumen. Do.
26 27 28			291	No albumen; dark-brown color; dog has lassitude; does not eat bread nor take milk, only a little meat. No albumen; dark-brown color; dog has lassitude; dog eats little. At the point of injection an abscess appears; urine acid; no
29 30 Apr. 2			270 295 280	albumén; yellow color. Urine acid; no albumen; yellow color; dog eats more. Urine acid; no albumen; yellow color; abscess broke. Urine normal; appetite almost normal.
Total.	$\left\{\begin{array}{c} 15.0 \\ 20.5 \end{array}\right.$			

<sup>&</sup>lt;sup>1</sup> By mouth.

2. See also Chlopin (Zts. Nahr. Genussm., 1902, v. 5, p. 241).

#### DOUBTFUL.

1. WINOGRADOW (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

# G. T. 667.

Trade names.—Quinolin Yellow; Quinolin Yellow, water-soluble.

Name under which it was offered on the United States market as a food color in 1907.—Chinolin Yellow O.

Scientific name.—Sodium salt of the sulphonic acid (chiefly disulphonic acid) of quinophthalone.

Discovered.—1882.

Shade.—Greenish Yellow. Offered by 1 out of 12 sources.

FAVORABLE.

Nothing.

#### UNFAVORABLE.

1. Chlopin (p. 205) on his own experiments reports this color as "suspicious." The experimental data are as follows:

Experimental data by Chlopin.

No. 1.

[1 gram=44 mg=31 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 19 20 21 22	Grams. 3.00	Kilos. 22. 6	cc. 730 720 749	Before experiment dog and urine normal. Urine dark brown; traces albumen; acid. Color of urine normal; no albumen. Do.

97291°-Bull. 147-12-10

<sup>&</sup>lt;sup>2</sup> Subcutaneously.

# Experimental data by Chlopin-Continued.

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Mar. 23 24 25 26 27 28 29 30	Grams. 0.75	Kilos. 21.4	cc. 750 693 725	Urine dark brown; no albumen. Dog eats nothing. Do. Dog eats nothing; eats poorly; albumen in urine. Dog eats nothing; rather much albumen. Do. Do. Dog eats nothing; traces of albumen.
Apr. 2 4 5 6 8 Total	1 3.00		710	Color of urine almost normal; insignificant traces of albumen. Urine yellow brown; traces of albumen; appetite normal. Urine yellow brown; no albumen. Urine and dog normal.

No. 2.

#### [1 gram=133 mg=83 grains.]

1903. Mar. 3 4 5 6	3. 00 3. 00 3. 00 2. 00	7.5	320 300 300	Before experiment dog quite well, and urine normal. Color dark yellow; no albumen; general condition normal. Color yellower than normal; acid; no albumen. Do.
Total	11.00			

<sup>1</sup> By mouth.

#### DOUBTFUL.

1. Winogradow (Zts. Nahr. Genussm., 1903, v. 6, p. 589) says it noticeably retards digestive action; is not indifferent.

# G. T. 670.

Trade names.—Vidal Black; Vidal Black S.

Scientific name.—Possibly the sulpho-hydro derivative of a polythiazin.

Discovered.—1893.

Shade.—Green. Not offered.

FAVORABLE.

Nothing.

UNFAVORABLE.

 Chlopin (p. 208) examined this color, and on his own experiments classes it as "very harmful." The experimental data are as follows:

Experimental data by Chlopin.

[1 gram=132 mg=92 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.					
1901. Dec. 29	Grams. 2	Kilos.		Before experiment dog quite normal; soon after taking dye vomited twice. General condition good; no vomiting; urine turbid, alkaline; no albumen.					

<sup>&</sup>lt;sup>2</sup> Subcutaneously.

# Experimental data by Chlopin-Continued.

Date.	Dose.	Welght.	24 hours' urine.	General condition of animal and urlne.
1901. Dec. 31	Grams.	Kilos.	cc.	Vomited soon after getting dye; appetite less; no albumen.
1902. Jan. 1 2 4 5	3 3 3	7	369 370	Urine turbid; scarcely noticeable blackish sheen; green with sulphuric and hydrochloric acid. Do. Vomited all day after taking dye; eats little; no albumen. Violent and prolonged vomiting and diarrhea; condition pitiable; further giving of dye stopped, so as not to kill the animal.
6-7 8 9-11 13				Vomiting and diarrhea continue. Vomiting and diarrhea stopped. Dog is livelier, and begin' to eat. Dog looks well; urine normal in color; acid; no albumen.
Total	17			

# G. T. 675.

Trade names.—Thiocatechin; Thiocatechin S. Shade.—Brown. Not offered.

FAVORABLE,

Nothing.

UNFAVORABLE.

Chlopin (pp. 210, 211) examined this color and classes it as "very poisonous."
 The experimental data are as follows:

# Experimental data by Chlopin.

[1 gram=128 mg=90 grains.]

Date.	Dose.	Weight.	24 hours' urine.	General condition of animal and urine.
1902. Feb. 13	Grams. 2.4	Kilos. 7.8	cc. 300	Before experiment dog and urine quite normal; 10 or 15 minutes after giving dye dog fell on one side, limbs extended, stomach drawn in; small and frequent convulsions; retching; then abundant vomiting, same color as dye; soon after vomiting dog got up and walked as if drunk; hind legs tend to collapse; sali-
14	2.0		290	vation.  A few minutes after giving dye, dog again fell as if in an epileptic fit; convulsions of the extremities, which soon ceased, but the dog still lay stretched out, with open eyes, which reacted to light; tongue hanging out to one side; after 10 or 15 minutes vomiting began; dog still lying on one side, assumed a more normal attitude; 15 minutes later dog got up, walked as if
15 16 18	0.5			drunk; poor control over hind legs. Vomits at once after getting dye, but remained standing. Dog appears depressed, but eats. Dog quite well; urine usual normal color; no albumen.
Total	4. 9			

# G. T. 689.

Trade names.—Indigo; Indigo Pure BASF. Scientific name.—Indigotin. Shade.—Blue. Not offered.

#### FAVORABLE.

- 1. Permitted by Confectioners' List.
- 2. Permitted by the law of Italy.

#### UNFAVORABLE.

1. Fraenkel (p. 571): "However, pure Indigo, according to Kobert, is, in finely divided condition, a violent local irritant."

# G. T. 692.

Trade names.—Indigo Carmine; Indigo Extract; Indigotin.

Names under which it was offered on the United States market as a food color in 1907.—Indigo Carmine Powder IN; Indigotin; Indigotin A.

Scientific name.—Sodium salt of Indigotin disulphonic acid or the free acid.

Discovered.—1740.

Shade.—Blue. Offered by 3 out of 12 sources.

#### FAVORABLE.

- 1. Schacherl (p. 1046): "No objection to its use."
- 2. Santori (Moleschott's Untersuchungen, 1895, v. 15, p. 41): A dog weighing 4,500 grams received 90 grams dye in 30 days; vomited twice during the examination; no change in weight; animal killed with chloroform; autopsy showed slight dull swelling in the epithelium and convoluted canals of the kidneys. This dosage amounts to 667 milligrams per kilo per day, or 467 grains per 100 pounds per day. Classes it as harmless.

## ALPHABETICAL INDEX OF TRADE NAMES OF COAL-TAR COLORS.

The following list of the trade names of coal-tar colors appearing in the foregoing compilation on physiological action is complete when supplemented by the list of 23 colors given on page 227; Green Table numbers in parenthesis.

Acetin Blue (599).

Acid Brown (138).

Acid Green (434, 435).

Acid Green ex. conc. (435).

Acid Green conc. (435).

Acid Green conc. VN (435).

Acid Green conc. 780 (435).

Acid Fuchsin (462).

Acid Magenta (462).

Acid Magenta powd. (462).

Acid Rosein (462).

Acid Rubin (462).

Acid Violet (467).

Acid Violet 6 B (467).

Acid Yellow (8).

Acid Yellow AT (94).

Acid Yellow D (88).

Acid Yellow G (8).

Acid Yellow G pat. (8).

Acme Yellow (84).

Alizarin Blue ABS (563).

Alizarin Blue S (563).

Alizarin Green (527).

Alphanaphthol Orange (85).

Alsace Green (394).

Amaranth (107).

Amaranth B (107).

Anilein (593).

Anilin Blue Sp. Sol. (457).

Anilin Orange (2).

Anilin Purple (593).

Anilin Red (448).

Anilin Yellow (4).

Anilin Yellow S (4).

Anthracene Green (527). Anthracene Blue S (563). Archil Substitute V (28).

Archil Substitute V (28)

Atlas Orange (86). Auramin (425).

Auramin O (425).

Aurantia (6).

Aureolin (659).

Aurin (483).

Azalein (408). Azarin S (70).

Azin Blue, spirit soluble (599).

Azo Acid Rubin (103).

Azo Acid Yellow (92).

Azo Blue (287). Azo Flavin (92).

Azo Fuchsin G (93).

Azo Rubin (103).

Azo Rubin A (103). Azo Rubin S (103).

Bavarian Blue DBF (479). Bavarian Blue DSF (478).

Bengal Blue (602). Bengal Green (427). Benzoflavin (530).

Benzopurpurin 4B (277). Betanaphthol Orange (86).

Biebrich Scarlet (163).

Bismarck Brown (197). Bismarck Brown R (201).

Bitter-almond-oil Green (427).

Bleu de Nuit (457).

Bleu Lumière (457). Bleu Marine (480).

Bordeaux B (65).

Bordeaux B L (65). Bordeaux R ext. (65).

Bordeaux S (107).

Brilliant Black B (188).

Brillant Cotton Blue, greenish (479).

Brilliant Green (428).
Brilliant Orange (13)

Brilliant Orange (13). Brilliant Scarlet (106).

Brilliant Scarlet 4R (106). Brilliant Yellow (5).

Brilliant Yellow S (89).

Butter Yellow (16). Canary Yellow (425).

Carbazotic Acid (1).

Cardinal 3B (103).

Carminaph (11). Carmoisin (103).

Carmoisin B (103).

Cerasin (102).

Cerasın Orange I (11).

China Blue (480).

Chlorin (394).

Chrome Violet (593).

Chrysamin R (269).

Chrysaurein (86). Chryseolin (84).

Chrysoidin Crystals (17, 18, 41).

Chrysoidin R (17, 18, 41).

Chrysoidin Y (17, 18, 41).

Chrysoin (84).

Chrysoin REZ (84).

Cinnamon Brown (197).

Citronin (4). Claret Red (65).

Claret Red RZ (103, 106, 107).

Cœrulein S (527).

Cochineal Red A (106).

Congo Red (240).

Cotton Blue (480).

Cotton Blue R (639). Cotton Red 4B (277).

Coupier's Blue (600).

Crocein Orange (13).

Crocein Orange 10234 (13).

Crocein Orange G (13).

Crocein Scarlet (106). Crocein Scarlet 3B (160).

Crocein Scarlet 4BX (106). Crocein Scarlet 7B (169).

Crocein Scarlet 8B (169).

Curcumin (89). Curcumin S (399).

Dahlia (450, 451).

Dark Green (394).

Diamond Green B (427).

Dianthin G (516). Dinitro-cresol (2).

Dinitrosoresorcin (394).

Diphenylamin Blue (479).

Diphenylamin orange (88).

Direct Gray (576). Direct Violet (451).

Emerald Green (428).

Emerald Green cryst. (428).

English Brown (197).

Eosin (512). Eosin A (512).

Eosin A ex. (512).

Eosin B (512).

Eosin bluish (517).

Eosin G G F (512).

Harmalin (448).

Eosin J (512, 517). Eosin 10 B (521). Eosin 3J & 4J ext. (512). Eosin Y (512). Eosin Yellowish (512). Erika B (78). Erythrobenzin (448). Erythrosin (517). Erythrosin B (517). Erythrosin D (517). Erythrosin G (516). Erythrosin yellow shade (517). Ethyl Green (428). Fast Blue (477). Fast Blue B, spirit soluble (599). Fast Blue R and 3R (601). Fast Blue R, spirit soluble (599). Fast Blue 2R, B, and 6B (601). Fast Blue R, 2R, and 3R (639). Fast Brown G (138). Fast Green (427). Fast Green J (428). Fast Green O (394). Fast Myrtle Green (394). Fast Navy Blue R (639). Fast Navy Blue RM and MM (639). Fast Ponceau B (163). Fast Red (102, 105). Fast Red C (103). Fast Red D (107). Fast Red EB (107). Fast Violet (620). Fast Yellow (4, 8, 9, 88). Fast Yellow extra (8). Fast Yellow G (8). Fast Yellow 053 (8). Fast Yellow R (9). Fine Blue (457). Fuchsiacin (448). Fuchsin (448). Fuchsin cryst. (448). Fuchsin S (462). Gallocyanin DH and BS (620). Gallocyanin RS, BS, and D (620). Gentian Blue 6B (457). Gentiana Violet (649). Gentianin (649). Gold Orange (86). Gold Yellow (84). Golden Yellow (2, 3). Gray R and B (602).

Green E (428).

Green 087 (428).

Guinea Green (433).

Helianthin (87). Helvetia Blue (479). Hessian Blue (457). Hofmann Violet (450). Hydrazin Yellow (94). Imperial Scarlet (163). Imperial Yellow (6). Indigen D and F (599). Indigo (689). Indigo Carmine (692). Indigo Carmine powd. IN (692). Indigo extract (692). Indigo pure BASF (689). Indigotin (692). Indigotin A (692). Indisin (593). Indisin R and B (601). Indophenol (572). Indophenol white (572). Indulin opal. (599). Indulin sol. (601). Indulin, spirit soluble (599). Indulin 3B opal. (599). Indulin 6B opal. (599). Indulin 3B sp. sol. (599). Indulin 6B sp. sol. (599). Indulin 3B (601). Indulin 6B (601). Iodeosin B (517). Iodeosin G (516). Iodin Green (459). Iodin Violet (450). Jaune Acide (4, 8). Jaune Acide C (4). Jaune Naphthol (3). Jaune d'Or (3). Jaune d'Orient (516). Jaune Soleil (399). Kaiser Yellow (6). Kermesin Orange (97). Leather Brown (197). Leather Yellow (532). Lemon Yellow (4). Light Green S F bluish (434). Light Green S F yellow shade (435). London Blue extra (480) Magdala Red (614). Magenta (448). Magenta F A B S Red (448). Magenta Powder A (448). Maize (399). Malachite Green (427). Malachite Green B (427-428).

Malta Gray (576).

Manchester Brown (197).

Manchester Brown EE (201).

Manchester Yellow (3).

Mandarin G R (97).

Mandarin G ext. (86).

Martius Yellow (3).

Mauve (593).

Mauve Dye (593).

Mauvein (593).

Meldola's Blue (639).

Metanil Yellow (95).

Methyl Blue water soluble (478).

Methyl Blue for silk MLB (478).

Methyl Blue O (479).

Methyl Violet (451).

Methyl Violet B (451).

Methyl Violet BB ext. (451).

Methyl Violet 3 BD (451).

Methylene Blue (650).

Methylene Blue A ext. (650).

Methylene Blue B and BG (650).

Methylene Blue B D (650).

Methylene Blue BB extra (650).

Methylene Blue BB extra D (650).

Methylene Gray (576).

Methylene Green G. conc. ext. (651).

Naphthalene Pink (614).

Naphthalene Red (614).

Naphthalene Rose (614).

Naphthol Black B (188).

Naphthol Black BDF (188).

Naphthol Blue R & D (639).

Naphthol Green (398).

Naphthol Green B (398).

Naphthol Orange (85).

Naphthol Red S (107).

Naphthol Yellow (3, 4).

Naphthol Yellow L (4).

Naphthol Yellow S (4,5).

Naphthol Yellow SLOZ (4, 86).

Naphthylamin Yellow (3).

Naphthylene Yellow (2).

Navy Blue B (478).

New Coccin (106).

New Gray (576).

New Green (427).

New Methylene Gray (576).

New Red L (163).

New Victoria Green (427, 428).

New Yellow (88).

New Yellow L (8).

Nicholson Blue (477).

Night Green (459).

Nigrosin (576).

Nigrosin sol. (602).

Nigrosin sp. sol. (600).

Nitrodiphenylamin (6).

Nitromethylene Blue (651).

Oil Black (600).

Oil Orange 7078 (11).

Oil Yellow (16).

Opal Blue (457).

Orange (86).

Orange I (85).

Orange II (86).

Orange III (87).

Orange IV (88).

Orange A (86).

Orange A extra (86).

Orange Brown (17, 18, 41).

Orange A 1201 (86).

Orange B (85).

Orange extra (86).

Orange G (14).

Orange GG (14).

Orange GG Crystals (14).

Orange GRX (13).

Orange GS (88).

Orange GT (43).

Orange M (88).

Orange MN (95).

Orange N (43,88).

Orange O 27 (85).

Orange O (43).

Orange R (15, 55, 97).

Orange RN (43).

Orange RZ (85).

Orange T (97).

Orange 2 R (97).

Orange Y (86).

Orcellin No. 4 (102).

Paris Violet (451).

Phenylene Brown (197).

Philadelphia Yellow G (532).

Phloxin (521).

Phloxin TA (521).

Phosphin (532).

Picric Acid (1).

Pistachio (435).

Polychromin (659).

Pomona Green (459).

Ponceau B (163).

Ponceau 4 GB (13).

Ponceau 4 RB (160).

Ponceau G and GR (55).

Ponceau R (55).

Ponceau 3 RB (163).

Ponceau 2 G (15). Ponceau 2 R (55). Ponceau 6 RB (169). Primula (450). Primulin (659). Printing Blue (599). Pure Soluble Blue (480). Pyoctanin Aureum (425). Pyoctanin (451). Pyrosin B (517). Pyrosin J (516). Quinolin Yellow, water-soluble (667). Quinolin Yellow (667). Rauracienne (102). Red (107). Red Violet 5 R extra (450). Resorcin Yellow (84). Resorcein O 275 (84). Rhodamin (504). Rhodamin B (504). Rhodamin O (504). Rhodamin B extra (504). Rhodamin G and G extra (502). Roccellin (102). Rose Bengal (520). Rose Bengal AT (520). Rose Bengal G (520). Rose Bengal N (520). Rosein (448). Rosolic acid (483). Rubianite (448). Rubidin (102). Rubin (448). Rubin S (462). Russian Green (394). Saffron Substitute (2). Saffron Yellow (3, 4). Safranilin (504). Safranin (584). Safranin AG, AGT, and OOF. (584). Safranin Conc. (584). Safranin extra G (584). Safranin FF extra (584). Safranin GOO. (584). Safranin S. (584). Safranin SP. (584). Safranin T. (584). Scarlet (55). Scarlet L. (106). Sloelin (600). Sloelin RS. and BS. (601). Solferino (448). Solid Yellow (4).

Soluble Blue (480).

Spirit Black (600).

Spirit Blue (457). Soluble Blue 8 B. (479). Soluble Blue 10 B. (479). Soluble Blue XL. (479). Succinic (4). Sudan I (11). Sudan Red (614). Sulphin (659). Sulphin Yellow (4). Sulphonaphthol Acid Yellow (4). Sultan Red 4 B. (277). Sun Yellow (399). Tartrazin (94). Thio Catechin (675). Thio Catechin S. (675). Toluidin Blue O. (654). Tropæolin D. (87). Tropæolin G. (95). Tropæolin O (84). Tropæolin OO (88). Tropæolin OOO (85). Tropæolin OOO No. 2 (86). Tropæolin R (84), Ursol D. (574). Ursol DD. (574). Ursol P. (574). Vert Diamant (427). Vert Lumière (459). Vesuvin (197). Vesuvin B. (201). Victoria Blue B. (488). Victoria Blue 4 R. (490). Victoria Orange (2). Victoria Yellow (2). Victoria Yellow Conc. Z. (95). Vidal Black (670). Vidal Black S. (670). Violein (593). Violet de Methylanilin (451). Violet R. (450). Violet RR. (450). Violet 5 R. (450). Water Black (166). Water Blue 6 B extra (480). Water Blue (480). Xanthin (532). XL Soluble Blue (479). Xylidin Red (55). Xylidin Scarlet (55). Yellow Corallin (483). Yellow FY. (4). Yellow MXX Conc. (95). Yellow W (9).

Yellow WR. (89).

# X. DOSAGE AND SYMPTOMS.

## CONFECTIONERS' LIST AS A BASIS FOR A RULE.

Considering the Confectioners' List of 1899 as a correct guide as to which colors are harmful and which are harmless, the attempt has been made to determine how far dosage and the corresponding physiological effects may serve as a guide in determining which colors, other than those enumerated in either portion of the Confectioners' List, are harmful or harmless. (See p. 48.)

To this end the available literature has been searched and classified, and wherever it was possible to arrive at any conclusion as to the actual dose or the average dose over a stated period of time, and the corresponding physiological observations, these data have been separated and brought together for the purpose of making comparisons and deductions therefrom.

It was thought that the literature would show that if a dog or other animal is killed by a certain given amount of color per 100 pounds of body weight of the animal that such color is always harmful; that if untoward effects, such as vomiting, diarrhea, weakness, and general depression, are caused by more than a certain weight of color per 100 pounds body weight of the animal, such color is always regarded as harmless.

The classification of the available literature and the conclusions therefrom are as follows:

Of the 33 coal-tar colors listed as harmless in the Confectioners' List, 10, namely, G. T. 4, 9, 55, 65, 85, 103, 105, 107, 448, and 462, have been tested on humans, while the conclusion as to the remaining 23 is reached by the effects observed on dogs alone. However, contradictory statements are recorded in the case of No. 9, and none of these tests was of long-continued duration, but, on the contrary, in many cases the time covered was exceedingly short, and the conclusions deduced are, therefore, not necessarily final nor correct. It should be further noted that Nos. 95 and 106, reported as nonpoisonous to humans, are in the harmful section of this Confectioners' List.

I. Those colors which produced no effect are as follows (the number of grains given is the amount administered per 100 pounds body weight; where the data permitted, the number of days' duration of the experiment is also given:

	*	Gr	ains.
5.	Brilliant Yellow S		532
13.	Ponceau 4 GB		113
521.	Phloxin		100
521.	Phloxin		300

II. Those colors which in some cases produced no effect, and in others produced effects, are as follows:

4. Naphthol Yellow S:

395 grains produced diarrhea and no albuminuria.

292 grains produced albuminuria.

116 grains for 25 days produced no effect.

25 grains for 2 weeks, on alternate days, produced no effect.

8 and 9. Fast Yellow Y and R:

173 grains for 3 weeks produced no effect.

53 grains produced albuminuria.

55. Ponceau 2 R.

582 grains killed.

198 grains produced no effect.

65. Fast Red B:

143 grains for 145 days produced no effect.

137 grains for 145 days produced no effect.

98 grains for 145 days produced no effect.

68 grains produced albuminuria.

38 grains produced albuminuria.

103. Azorubin S:

143 grains for 145 days produced no effect.

137 grains for 145 days produced no effect.

98 grains for 145 days produced no effect.

70 grains produced diarrhea.

287. Azo Blue:

233 grains produced no effect.

166 grains produced albuminuria.

III. Those that produced only a slight disturbance which was regarded as negligible:

28. Archil Substitute V:

301 grains produced vomiting and albuminuria.

127 grains produced vomiting and albuminuria.

105. Fast Red E:

70 grains produced diarrhea.

166. Wool Black:

117 grains produced albuminuria.

240. Congo Red:

192 grains produced albuminuria.

269. Chrysamin R:

433 grains produced albuminuria.

361 grains produced albuminuria.

394. Dinitrosoresorcin:

139 grains produced loss of appetite.

121 grains produced albuminuria.

From Class I above it would appear—

(a) That if a dose of 113 grains per 100 pounds body weight produced no effect it is to be classed as harmless.

From Class II above it would appear—

(a) That if albuminuria is caused by as little as 38 grains in some cases, and no effect is produced by doses as large as 143 grains, the color is to be classed as harmless.

- (b) Also that if 198 grains produce no effect the color is to be classed as harmless, even if 582 grains in another case kill the animal.
- (c) That if small amounts produce albuminuria and larger amounts do not the color is to be classed as harmless.
- (d) That if small amounts produce diarrhea and larger amounts do not the color is to be classed as harmless.

From Class III it would appear that—

- (a) Albuminuria produced by as little as 117 grains is to be regarded as not harmful.
  - (b) Diarrhea produced by 70 grains is to be regarded as not harmful.
- (c) Vomiting and albuminuria produced by 127 grains are to be regarded as not harmful.

The colors classed as harmful in the Confectioners' List may be classified as follows:

I. Dogs.—Those colors that under certain conditions produce no noticeable effects on dogs, while under other conditions effects are observed, are classed as harmful colors:

# 1. Pieric Acid:

Dogs stand 5 grains daily for 18 days without effect.

18½ grains produced diarrhea.

20.4 grains killed.

## 2. Dinitrocresol:

140 grains caused vomiting but did not kill.

 $38\frac{1}{2}$  grains caused vomiting but did not kill.

38½ grains caused vomiting and did kill.

35 grains caused vomiting but did not kill. 31.6 grains caused vomiting but did not kill.

# 3. Martius Yellow:

51 grains do not kill, but produce weakness, diarrhea, and albuminuria.

50 grains kill.

8 grains do not kill, but produce weakness, diarrhea, and albuminuria.

#### 86. Orange II:

714 grains produced kidney irritation, thirst, and diarrhea.

333 grains kill, and produced diarrhea and albuminuria.

244 grains produced kidney irritation, thirst, and diarrhea.

36 grains produced no effect.

# 95. Metanil Yellow:

620 grains produce vomiting.

603 grains kill.

407 grains no effect.

371 grains kill.

104 grains produced albuminuria.

62 grains no effect.

41 grains no effect.

31 grains kill.

## 197. Bismarck Brown:

246 grains produced vomiting.

118 grains produced vomiting and general depression.

38½ grains daily for 30 days, no effect.

398. Naphthol Green B:

600 grains produced green urine and conjunctiva.

292 grains produced no effect.

128 grains produced green urine and conjunctiva.

# II. Those colors that have not been observed to give negative results, but which have caused certain effects:

## 11. Sudan I:

118 grains produced colored urine, vomiting, and albuminuria.

17 and 18. Chrysoidin Y and R:

79 grains produced albuminuria.

74 grains for 30 days produced no albuminuria, but a loss of 12½ per cent of body weight.

88. Diphenylamin Orange:

216 grains produced albuminuria.

128 grains produced phenol in urine and albuminuria.

138. Fast Brown G:

237 grains produced diarrhea and loss of appetite.

218 grains produced diarrhea.

## III. Rabbits:

# 1. Picric Acid:

24.5 grains for 90 days, no effect.

45 grains for 19 days kill.

## 86. Orange II:

292 grains, no effect.

933 grains kill.

## 95. Metanil Yellow:

216 grains, no effect.

## IV. Humans:

### 1. Picric Acid:

8.3 grains no effect.

13.8 grains no effect.

Invalids and children can not stand this color.

# 2. Dinitrocresol:

42 grains kill.

#### 86. Orange II:

1½ grains, no effect.

3 grains, headache, vertigo, dryness of throat, and poor general condition.

# 95. Metanil Yellow:

 $1\frac{1}{2}$  grains, no effect.

3 grains, no effect.

# From Classes I and II above, it would seem to appear—

(a) That if a dog is killed the color is harmful, even though it take as much as 603 grains, or as little as 20.4 grains per 100 pounds body weight to kill.

(b) That if albuminuria is produced in dogs by as little as 79 grains, or as much as 128 grains, the color is harmful.

(c) That if diarrhea is produced in dogs by as much as 218 grains, or as little as 50 grains, the color is harmful.

(d) That even if a dog can take as much as 407 grains, the color may be harmful.

# From Class III it would appear-

(a) That even if rabbits can stand as much as 292 grains, or as little as 24.5 grains, the color is harmful.

# From Class IV it would appear—

(a) That if 42 grains kill a human, the color is harmful.

(b) That even if humans can stand as much as 3 grains without untoward effect, the color is harmful.

# The conclusions that may be drawn from these data are:

1. If a dog is killed by-

- (a) 603 grains per 100 pounds body weight, the color may be harmful.
- (b) 582 grains per 100 pounds body weight, the color may be harmless.

2. If a dog can bear without effect-

- (a) 407 grains per 100 pounds body weight, the color may be harmful.
- (b) 198 grains per 100 pounds body weight, the color may be harmless.

3. If albuminuria is produced in a dog by-

- (a) 38 grains, or 143 grains per 100 pounds, body weight the color may be harmless.
- (b) 79 grains, or 128 grains per 100 pounds body weight, the color may be harmful.
- 4. If diarrhea is produced in a dog by-
  - (a) 70 grains per 100 pounds body weight, the color may be harmless.(b) 50 grains per 100 pounds body weight, the color may be harmful.
- 5. If vomiting and albuminuria are produced in a dog by 127 grains per 100 pounds body weight, the color is not necessarily harmful.
- 6. If small amounts of color produce in a dog diarrhea or albuminuria, and larger amounts do not, the color may be harmless.
- 7. Even though rabbits can withstand 292 grains per 100 pounds body weight, the color is not necessarily harmless, but may be harmful.
  - 8. If 42 grains kill a human, the color is harmful.
- 9. If a human can withstand 3 grains without effect the color is not necessarily harmless, but may be harmful.
- 10. If a human can not withstand 3 grains, even though it can withstand 1½ grains, the color is not necessarily harmless, but may be harmful.

# Lehmann (Methoden der praktischen Hygiene, 1890, p. 545) says:

I regard such substances as harmful to health which when fed to a sound dog in doses of a few decigrams per day produce at once, or after a few repetitions of the dose, disturbances in the health of the dog; on the other hand, dyes which in doses of from one to several grams can be taken for weeks on end without causing any disturbance or only slight intestinal disturbances or a slight and passing albuminuria can be regarded as harmless. It should never be forgotten that a few milligrams of a coaltar color dyes very strongly and it is not easy—even by most extraordinary use of colored objects, e. g., by children—that more than milligrams, at most centigrams, of dye can be introduced into the human stomach. In extremely large doses many substances, for example, all our condiments, are naturally harmful.

Of 65 dogs weighed and experimented on by Chlopin the average weight was 9.2 kilograms, or 20.3 pounds; almost half the dogs (30) weighed between 6 and 11 kilos.

To adapt the rules of Lehmann to a basis of grains per 100 pounds of body weight, assuming the average weight of a dog to be 20 pounds,

it is only necessary to multiply the weight of color by 5 and by 15.432. Doing so, the following data are obtained:

A. Harmful colors produce bad effects in doses of a "few" decigrams, i. e., multiples of a half gram; the word "few" is not sharply defined, but the rule means a "few" times 7.7 grains.

B. Harmless colors produce, when continuously fed, no bad effect in multiples of 10 of the doses of A, above, i. e., multiples of 77 grains.

C. Slight diarrhea and slight, temporary albuminuria are not to count against the color

Applying these rules to the preceding colors discloses that they were apparently not so used, in selecting the harmless and harmful colors in the Confectioners' List.

#### LEHMANN'S RULES.

The Lehmann rules as just interpreted were applied to the data just given. As a result of such application the conclusion is reached that of the 15 colors classed as harmless (foregoing classes I, II, and III), 6 would be classed as doubtful by the Lehmann rules, namely:

13. Ponceau 4 GB,

65. Fast Red B,

8 and 9. Fast Yellow Y and R,

103. Azorubin S,

55. Ponceau 2R,

105. Fast Red E,

and the remaining 9 would have been classed as harmless.

Of the 11 classed as harmful (foregoing classes I, II, III, and IV), 2 would have been regarded as harmless, namely:

398. Naphthol Green B,

138. Fast Brown G,

3 would have been classed as doubtful, namely:

197. Bismarck Brown,

88. Diphenylamin Orange,

11. Sudan I,

and the remaining 6 would have been classed as harmful.

# SANTORI'S WORK AS A GUIDE TO A RULE.

Santori regards the following six dyes as harmless: 457 (188), 467 (74), 477 (129.5), 480 (156), 599 (64), 692 (467).

The first is the Green Table number and the bracketed figure the average number of grains per 100 pounds per day for 30 days.

In the case of Nos. 457 and 599 the animal was wholly normal throughout the test and the autopsy showed only normal conditions.

In the case of No. 477 the only disturbance was colored feces and the autopsy showed only normal conditions.

In the case of No. 480 the only disturbance was colored feces, but the autopsy showed a pea-green kidney.

In the case of Nos. 467 and 692 there was only vomiting; in the case of No. 467 the autopsy showed everything normal; whereas in the case of No. 692 the autopsy disclosed a swollen kidney.

Therefore, according to Santori, a dye that causes colored feces, even with colored kidney, is harmless. Also a dye that causes vomit-

ing and swollen kidney is harmless. On the other hand, a dye (572) which produces no change or symptom observable during life, but fatty degeneration of the liver is shown at the autopsy, is a harmful dye. This lack of conformity makes the relation between dosage, symptoms, and harmfulness or harmlessness more confusing and perplexing.

# YOUNG'S RULE.

It must be remembered that smaller amounts of drugs, and, therefore, of coal tar colors, effect children as a rule than are effective upon adults. Taking Young's rule as a guide, it appears that generally the effective dose for a 3-year-old child is one-fifth the effective adult dose; for a 4-year-old child one-fourth; for a 6-year-old child one-third; for an 8-year-old child two-fifths and for a 12-year-old child one-half the effective adult dose. All of this should be taken into account in drawing conclusions from experiments as to the harmlessness of any coal-tar dye upon humans. Very little attention has, however, been paid to this aspect of the matter in spite of the fact that colored foods, confectionery, pastry, beverages, and the like are partaken of by children and in many cases such articles are prepared for the sole or particular consumption of the very young.

From the foregoing data it seems clear that deductions as to the harmlessness or harmfulness of coal-tar dyes when administered to dogs, and not based upon autopsies, are not final nor conclusive as to the effect upon the dog. The extent to which such deductions are correctly transferable to humans is likewise not established.

## XI. OIL-SOLUBLE OR FAT COLORS.

Oil-soluble colors are used for coloring fats, such as butter, oleomargarine, edible oils, and the like; of the coal-tar colors the oilsoluble colors are chemically nonsulphonated azo-colors.

The nonsulphonated azo-colors which have been physiologically examined are:

- 11. Sudan I (anilinazo-b-naphthol) (2).
- 16. Butter Yellow (anilin-azo-dimethylanilin).
- 17. Chrysoidin Y (anilin-azo-m-phenylene-diamin) (2).
- 18. Chrysoidin R (anilinazo-m-tolylene-diamin) (1).
- 41. Chrysoidin R (o-toluidin-azo-m-tolylene-diamin).
- 197. Bismarck Brown (m-phenylene-diamin-disazo-m-phenylene-diamin) (4).
- 201. Manchester Brown (m-tolylene-diamin-disazo-m-tolylene-diamin) (2).

The numbers preceding the trade names are the Green Table numbers; the scientific names appear in parentheses; and the number of dealers offering the colors on the United States market in the summer of 1907, out of a possible 12, appear after the scientific name, also in parentheses.

For all but No. 201 contradictory statements occur in the compiled literature; for No. 201 only unfavorable reports were found.

There were on the United States market in the summer of 1907 the following oil-soluble colors:

- 10. Sudan G (anilin-azo-resorcin) (1).
- 11. Sudan I (anilin-azo-b-naphthol) (2).
- 49. Sudan II (xylidin-azo-b-naphthol) (1).
- 60. Carminaph Garnet (a-naphthylamin-azo-b-naphthol) (1).

Of these No. 11 had been reported on contradictorily, and the remaining three had not been reported on at all. In addition to these the following three oil-soluble colors, not listed in the Green Tables and not reported on in literature, were wanted:

- 1. o-Toluidin-azo-b-naphthylamin (which does not seem to be described in literature in any way).
  - 2. Amidoazo-toluol.
  - 3. Anilin-azo-b-naphthylamin.

So that out of a total of seven oil-soluble colors on the United States market in the summer of 1907 only one had been examined physiologically and that with contradictory results. As before stated, these oil-soluble colors all belong to the class of nonsulphonated azo-colors.

Fraenkel (p. 575) says: "When, however, the azo-colors contain no sulpho-group (i. e., are nonsulphonated) they are poisonous. Thus for example, Bismarck Brown \* \* \* Sudan I." Yet meta-nitrazotin, a nonsulphonated color not in the Green Tables, and probably not upon the market anywhere, and which is meta-nitranilin-azo-b-naphthol, is according to Weyl nonpoisonous.

Fraenkel (p. 575) also stated:

The fact that the monazo colors examined by Cazeneuve and Lépine are harmless, as above stated, is equally explained by the constitution of these substances. Those two investigators examined (omitting the trade names) a-naphthylamin-sulphoacid-azo-a-naphthol-acid, a-naphthylamin-sulphoacid, a-naphthylamin-azo-b-naphthol-disulphoacid, xylidin-azo-b-naphthol-disulphoacid, sulphanilicacid-azo-a-naphthol, Amido-azo-toluene-disulphonicacid.

These substances are all sulphoacids and the sulpho-groups here effect the depoisoning of the original substance.

Examining this statement it therefore appears that in Fraenkel's opinion, at least, a-naphthylamin-azo-a-naphthol, a-naphthylamin-azo-b-naphthol, xylidin-azo-b-naphthol, anilin-azo-a-naphthol, and amido-azo-toluol are in and of themselves poisonous substances, and are rendered nonpoisonous by sulphonation. Diligent search through the literature has failed to uncover any original communications of investigators to that effect.

With but one known exception all the nonsulphonated azo-colors are regarded by Fraenkel as poisonous. The colors 17, 18, 41, 197, and 201 on page 159 are not used as oil-soluble colors and therefore form no comparative basis for judging oil-soluble colors except that

both are nonsulphonated azo-colors; in Fraenkel's opinion, however, such a comparison is wholly justified and proper.

Excluding now 17, 18, 41, 197, and 201 for the purposes of comparison, the following tabulation is made:

Oil-soluble nonsulphonated azo-colors on the market, 1907, deemed poisonous by Fraenkel.

Oil-soluble colors.	On United States market.	Fraenkel calls poisonous.
anilin-azo-resorcin anilin-azo-b-naphthol xylidin-azo-b-naphthol a-naphthylamin-azo-b-naphthol o-toluidin-azo-b-naphthylamin amido-azo-toluol anilin-azo-b-naphthylamin a-naphthylamin-azo-a-naphthol anilin-azo-a-naphthol	x x x x x x	x x x

That is, the only ones on the United States market concerning which no expression of specific opinion is to be found in Fraenkel are the following three: anilin-azo-resorcin, toluidin-azo-b-naphthylamin, and anilin-azo-b-naphthylamin, and nothing is published on the physiological action of any of them specifically. The probability, however, would seem to be that they are not harmless, and until their harmlessness is positively established their exclusion from the permitted list of coal-tar colors for use in foods seems to be the only safe and proper course to be followed. The physiological action of so-called "Butter Yellow" No. 16 of the Green Tables (see p. 85), merely strengthens this conclusion.

# XII. RULES AND REASONS FOR SELECTING THE SEVEN COLORS PERMITTED BY F. I. D. 76.

# STATEMENT OF RULES.

In view of the confusion and uncertainty disclosed in the foregoing literature relative to the physiological action of coal-tar colors, the difficulties in the way of making a list of coal-tar colors to be permitted in the coloring of food products are seen to be by no means slight. The following rules governing selection were in mind during the making of the list given in Food Inspection Decision No. 76, and the closeness with which they were followed is discussed on page 166.

Rule I. All colors which have not been physiologically tested either upon animals or man shall not be permitted for use in foods.

Rule II. All coal-tar colors which have been examined physiologically with contradictory results shall not be permitted for use in foods.

Rule III. All coal-tar colors which have been examined physiologically and have been declared to be of doubtful harmlessness shall not be permitted for use in foods.

Rule IV. Only those coal-tar colors whose chemical composition was definitely disclosed or otherwise ascertained, and which were on the United States market in the summer of 1907, and which have been examined physiologically and with no other than a favorable result shall, for the present, be permitted for use in foods.

# ANALYSIS OF THREE RECOMMENDATIONS MADE TO THE DEPARTMENT OF AGRICULTURE.

These rules were formulated as a guide in view of the divergent opinions expressed in three different recommendations to the Department of Agriculture. One of these recommendations suggested that permitted colors be designated by nine titles. Comparison of these titles with the Green Tables and with the tabulated survey of the unfavorable, favorable, and contradictory literature corresponding to the Green Table numbers (p. 63) discloses the following facts:

Comparison of nine suggested color titles with corresponding Green Table numbers and the reports on the same.

Name.	Green Table No.	Un- favor- able.	Fa- vor- able.	Con- tra- dic- tory.	No re- port.1	Name.	Green Table No.	Un- favor- able.	Fa- vor- able.	Con- tra- dic- tory.	No re- port.1
Chrysoidin	17 18			x x		Ponceau	147 148				x x
Tropæolin	84 85 86		х	x x x			150 160 163 169		x	x x	x
	87 88 95			X X X		Bordeaux	65 107 157		x x		x
Azoflavin Roccellin Ponceau	92 102 13		x x	X			170 171 198				X X X
	15 44 55 56			x x	x	Biebrich Red (?) Sulphonated Fuchsin	244 20 462		x		X
	57 108 114				X X X	Naphthol Yellow S	4		x		
	146				x	Total	36	0	8	13	15

<sup>1</sup> Of physiological tests in literature compiled.

Therefore, under 9 titles 36 different chemical individuals would be placed upon the permitted list, of which only 8, or less than 25 per cent, have been examined physiologically with only favorable results, and 28, or more than 75 per cent, had either not been examined at all physiologically or with contradictory results.

The 8 of these chemical individuals examined with only favorable results, and the number of sources out of a possible 12 offering them on the United States market in the summer of 1907, are as follows:

Colors reported on favorably and number of dealers handling same.

Green Table No.	Sources offering.	Green Table No.	Sources offering.
1 4 65 85 92	10 2 2 2 0	102 107 169 462	0 7 1 2

1 The italicized Green Table numbers are those of the permitted list of Food Inspection Decision No. 76.

Another recommendation suggested the permissive use of 42 entries in the Green Tables, which are tabulated below in the same manner as the suggestions in the preceding recommendation:

Comparison of 42 recommended Green Table numbers with reports in the literature.

Green table No.	Un- favor- able.	Favor- able.	Equiv- ocal.	No report.1	Green table No.	Un- favor- able.	Favor- able.	Equiv- ocal.	No report.1
4 8 17		х	x		427 434	x		x	
50 53			x	x x	448 451 456		1	x x	x
54 55 65		x	х	X	457 462 477		X X	x	
85 86 89 92		X X X	x		478 479 480 504	x x		x x	
94 102 103	х	x x			512 513 514		х		X X
107 138 163		x	X		516 517 518	х	х		x
166 240 269		x x	x		562 601			х	x
287			x		Total	5	14	15	8

1 Of physiological tests in compiled literature.

Applying the conclusions hereinbefore reached to these entries, it is found that 14 out of the 42 colors recommended, or exactly one-third, had been examined physiologically with only favorable results, and the remaining two-thirds had been either examined physiologically with only unfavorable or with conflicting results, or had not been examined at all.

The 14 chemical individuals examined with favorable result and the number of sources out of a possible 12 offering them on the United States market in the summer of 1907 are as follows:

Colors reported	on favorably	and number o	f dealers	handling same
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Green table No.	Sources offering.	Green table No.	Sources offering.
1 4	10	107	7
65	2	166	0
85	2	240	1
89	1	462	2
92	0	477	0
102	0	512	3
103	6	577	5

<sup>&</sup>lt;sup>1</sup> The italicized Green Table numbers are those of the permitted list of Food Inspection Decision No. 76.

The third of these recommendations suggested 27 different chemical individuals for permissive use; these are tabulated below in the same manner as the suggestions in the preceding recommendations.

Twenty-seven colors recommended for use and reports in the literature on the same.

Un- favor- able.	Favor- able.	Equiv- ocal.	No report.1	Green table No.	Un- favor- able.	Favor- able.	Equiv- ocal.	No report.
	x	X X		481 512 516	x	x		x
	x	x x		517 518 521		x		х
x	x	x		584 601 602	x		x x	
	x	x	x	692 Total	4	10	10	3
	favorable.	favorable.  x  x  x  x  x  x  x	favor- able.	favor- able.	favorable.	favorable.         Pavorable.         Equity ocal.         report.         table No.         favorable.           x         x         512         x         512         x         516         x         517         x         518         x         518         x         518         x         521         x         532         x         x         601         x         601         x         x         602         x         602         x         693         x         693         x         693         x         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7         7	favorable.         Pavorable.         Equity ocal.         report.         table No.         favorable.         Pavorable.           x         x         512         x         x           x         516         x         x           x         517         x         x           x         521         x         x           x         532         x         x           x         601         x         x           x         602         x         x           x         650         x         x           x         693         x         x           x         x         7         7         7         7	favorable.         Pavorable.         Equiverable.         Indicate of the point.         table.         favorable.         Favorable.         Equiverable.         Sequence of the point.           x         x         512         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         x         <

1 Of physiological tests in literature compiled.

Therefore, applying the same method of drawing conclusions as in the case of the preceding two recommendations, it appears that 10 out of the 27 suggested chemical individuals had been examined physiologically with only favorable results, and the remaining 17 had either not been examined at all or with unfavorable or conflicting results. The 10 chemical individuals examined with favorable results, and the number of sources out of a possible 12 offering them on the United States market in the summer of 1907, are as follows:

The 10 recommended colors favorably reported on in the literature and the dealers handling same.

Green	Sources	Green	Sources
Table	han-	Table	han-
No.	dling.	No.	dling.
1 4 65 85 102 462	10 2 2 2 0 2	477 512 517 521 692	0 3 5 0 3

<sup>&</sup>lt;sup>2</sup> The italicized Green Table numbers are those of the permitted list of Food Inspection Decision No. 76.

All of the colors included in these three recommendations concerning which only favorable reports were found in the literature are given in the following table:

Recommended color	favorably	reported on	in the	literature.
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Green	Rec	Recommendations.			Dealers offer-	Green Table	Recommendations.			Dealers offer-	
Table No.	I.	II.	III.	Total.	ing.	No.	I.	II.	III.	Total.	ing.
1 4 65 85 89 92 102 103 1 107 166 169	x x x x	x x x x x x x x	x x x	3 3 1 2 3 1 2 1	10 2 2 2 1 0 0 6 7 0	240 462 477 512 517 521 692 Total .	x	x x x x x	x x x x x x x	1 3 2 2 2 2 1 1	1 2 0 3 5 0 3

<sup>&</sup>lt;sup>1</sup> The Italicized Green Table numbers are those of the permitted list of Food Inspection Decision No. 76.

This table is now rearranged to show—

I. The recommended colors not offered in the United States market in the summer of 1907.

Green Table No.	Recom- mended by—	Green Table No.	Recom- mended by-
92 102 166	2 3 1	477 521	2 1

## II. The recommended colors offered on the United States market in the summer of 1907.

Green Table No.	Recom- mended by—	Sources handling.	Green Table No.	Recom- mended by—	Sources handling.
1 4 65 85 89 103 107	3 3 1 1 2	10 2 2 1 6 7	169 240 462 512 517 692	1 1 3 2 2 2	1 1 2 3 5 3

<sup>1</sup> The italicized Green Table numbers are those of the permitted list of Food Inspection Decision No. 76.

It appears that out of 17 different chemical individuals suggested by these 3 recommendations jointly, 5 were not on the United States market in the summer of 1907, and 12 were on that market; also that of these 12, 4 were suggested by all 3 recommendations; 3 were wanted by only 2 out of the 3, and the remaining 5 were spoken for by only one. Examining all of the recommendations, it appears that out of a total of 79 recommendations 17 had been examined with only favorable results, and that the remaining 62 were either not examined or had been examined with unfavorable or conflicting results.

To add to the confusion the third recommendation mentioned suggested that the following Green Table numbers should not be permit-

ted for use in food products because they are harmful: 1; 2; 3; 6; 86; 95; 425; 451; 454; 480; 483; 484; 487; 488; 490; 571; 584; 599; 620; 624; 650. The same paper, however, recommended that Nos. 584 and 650 be permitted for use in food products; that No. 86, suggested by both the other recommendations, be prohibited; and that Nos. 95, 451, and 480, suggested by one or the other of the remaining two recommendations, be likewise prohibited for use in food products. It must be clear to everyone that these three recommendations are not of one mind as to (1) the colors which are harmless; (2) the colors which are harmful; (3) the colors which are used or are useful in food coloring; and (4) the colors which should be used in food coloring. Such a state of affairs fully justifies rules of the scope and intent of those formulated at the beginning of this section for making a selection of permissible food colors.

## PROCESS OF ELIMINATION.

From the foregoing data it appears that out of 80 different chemical individuals on the food-color market of the United States in 1907, only 16 had been examined physiologically with a favorable result (see p. 64).

These 16, arranged according to the shade produced, are as follows (numbers preceding the names are the Green Table numbers; the numbers in parentheses show the number of sources desiring the color; the italicized Green Table numbers are the ones finally selected by Food Inspection Decision No. 76):

#### REDS.

- 65. Fast Red B (Alphanaphthylamin azo-R-salt) (2).
- 103. Azorubin S (Naphthionic acid azo NW acid) (6).
- 105. Fast Red E (Naphthionic acid azo-Schaeffer acid) (1).
- 107. Amaranth (Naphthionic acid azo-R-salt) (7).
- 169. Crocein Scarlet 7B (Amidoazotolutol-monosulphonic-acid-azo-betanaphthol-monosulphonic acid B) (1).
  - 240. Congo Red (Benzidin disazo-naphthionic acid) (1).
  - 462. Acid Magenta (Magenta trisulphonic acid) (2).
  - 512. Eosin A (Tetra-bromo-fluorescein) (3).
  - 517. Erythrosin (Tetra-iodo-fluorescein) (5).
  - 520. Rose Bengal (Dichlor-tetraiodo-fluorescein) (2).

#### ORANGE.

85. Orange I (Sulphanilic acid azo-alpha-naphthol) (2).

#### YELLOWS.

- 4. Naphthol Yellow (Dinitroalphanaphtholsulpho acid) (10).
- 89. Brilliant Yellow S (Sulphanilic acid azo-diphenylamin, sulphonated) (1).

#### GREENS.

- 433. Guinea Green B (Benzyldehyde + benzylethyanilin sulpho-acid) (1).
- 435. Light Green S F yellowish (Benzyldehyde+benzylethylanilin+sulphonation) (4).

BLUE.

692. Indigo Carmine (Indigo disulpho acid) (3).

Considering now the reds, and particularly 107, which, as the number appearing in parentheses after the scientific name indicates, was wanted by 7 sources out of the 12 drawn on, this being the most desired of all the reds, it would seem reasonable to believe that all the wants supplied by 65, 103, 105, 169, 240, and 462 would be covered by 107. Chemically 107 is closely allied to 65, 103, 105, and tinctorially it is likewise closely allied to 169, 240, and 462.

Nos. 512, 517, and 520 are chemically quite different from the other members of this group, and tinctorially they differ, being of a particularly brilliant shade, and tinctorially more powerful; 517 being desired by 5 out of the 12 sources drawn upon, and thus being the most desired of these three colors, was selected in the expectation that any work 512 and 520 could do in food products would be equally well done by 517. For reds, therefore, the choice fell upon 107 and 517.

Orange.—Only one color was wanted, and that by 2 out of the 12 sources, and this was placed in the permitted list.

Yellows.—The choice of No. 4, which was wanted by 10 out of the 12 sources, was made in the expectation that every purpose that 89 could serve in food products could be served also by 4.

Greens.—Tinctorially and chemically the two greens are very closely allied, and in view of the reasonableness of the expectation that 435 could do all the work of 433 as well, its choice was regarded as justified, especially as 435 was wanted by 4 out of the 12 sources, as against only one for 433.

Blue.—No. 692 being regarded as harmless by all, and being the only blue in the list, it was selected.

In this manner six out of the seven permitted colors were selected.

## REASONS FOR ADDING PONCEAU 3R.

An examination of the table on page 20 discloses the fact that among the reds desired four were azo-reds made from anilin derivatives as the first component, namely:

- 53. Xylidin-azo-alphanaphtholdisulpho acid.
- 54. Xylidin-azo-Schaeffer acid.
- 55. Xylidin-azo-R-salt.
- 56. Cumidin azo-R-salt.

Each of these was wanted by 1 source out of the possible 12. This was construed as an expression of a real need in the art of food coloring for a color of this class. The reason for such a need was not then, nor is it now, apparent, but the propriety of giving even a seeming need due consideration was regarded as justified. For only one of the four desired, namely, 55, could any specific references in the literature be

found, and, in fact, four references were found favorable thereto and three unfavorable thereto; thus eliminating 55 from consideration, leaving 53, 54, and 56.

The Austrian law of January 22, 1896 (see Lieber, p. 15), permits the use of azo colors derived from higher homologues of anilins, beginning with xylidin and sulphonated betanaphthols. As Nos. 53 and 54 were both derived from xylidin, it was regarded as safe to assume that they would probably be as objectionable as 55. This left 56 only to be considered, which color is derived from cumidin and R-salt. The cumidin portion satisfies the Austrian law and also satisfies the general law laid down in Fraenkel (p. 162), namely, that the greater the number of ring-methyls the less the toxic property of the resulting compound, because of the oxidation of these methyls to carboxyls in the animal system; the R-salt portion satisfies the general law laid down in Fraenkel, that the more highly sulphonated the less toxic a substance becomes, and therefore 56 is in the first portion of its composition of such a nature as to be less objectionable than 55, if 55 be objectionable.

This combination of facts, namely, the desirability of an anilin azo-red, the provision for diminishing the toxicity of 55 by the substitution of cumidin for xylidin therein, and the general provision in the Austrian food law, made it appear desirable and safe to take into account an anilin azo-red, and therefore to select 56 as probably the least objectionable, if it be at all objectionable, of the anilin azo-reds.

The full list of permitted colors was therefore extended to 7, as follows:

Red shades.—107. Amaranth; 56. Ponceau 3R; 517. Erythrosin. Orange shade.—85. Orange I.

Yellow shade.—4. Naphthol Yellow S.

Green shade.—435. Light Green S F yellowish.

Blue shade.—692. Indigo disulpho acid.

It should be noted that with respect to tetra-iodo-fluorescein, 517 of the Green Tables, no specific investigation, pronouncing it harmless or harmful, is described in the literature. No. 516, the diodo-fluorescein, is reported specifically adversely in the literature, especially by Chlopin; the Confectioners' List, the laws of Austria, the laws of France, the rather superficial examination of No. 517 by Grandhomme, and the statement in Fraenkel (p. 574), that it produces no disturbance, are all taken to apply to 517 and not to 516. The identification of the color under examination with 517 has not in all cases been satisfactorily exclusive, but it is believed that the differentiation of the harmful 516 by Chlopin makes all the other references cited pertinent to 517.

With respect to 435, it must be pointed out that 434, which is the methyl instead of the ethyl derivative, has only been regarded as suspicious, and one examination of 435, namely, that of Lieber (p. 144), does not appear to disclose anything which would positively exclude 435.

# QUALITY, CLEANLINESS, AND EFFICIENCY.

The justification for limiting the permitted colors to 6 out of 16, against which nothing unfavorable is contained in the literature and regarding which favorable statements are at hand, and the addition to these 6 of the seventh color is to be found in the very great variation in tinctorial quality, in percentage of coloring matter, in amounts of insoluble matter, both organic and inorganic, and in the amounts of organic matter not coloring matter, as well as in the large number of samples containing an amount of arsenic in excess of that permitted by the United States Pharmacopæia for the only coal-tar color therein mentioned, namely, Methylene Blue, and the varying amounts of heavy metals, such as copper, lead, and iron, mostly in excess of the limits permitted in the Pharmacopæia for various medicinal chemicals. This wide variation in quality and degree of cleanliness, all pointing to a very great difference in the care with which coal-tar colors offered for food purposes are prepared, render the conclusion safe that some control over food colors in respect to quality is desirable, necessary, and essential.

The efficiency of the 7 colors, 6 of which were selected from among the 16 considered for the purpose of making this list of permitted colors as being reasonably sure to be harmless, is evidenced by the fact that, although the addition of colors has been sought by persons interested in the food-coloring art, not one of the remaining 10 colors of those 16 has been so requested. In other words, the colors that the department has been requested to add to the permitted list were outside of the 16 colors which were on the markets of the United States in 1907, and were described in the literature in such a manner as to lead to the conclusion that they were probably not harmful.

# XIII. LISTS OF COLORS SUBSEQUENTLY RECOMMENDED BY INDIVIDUALS AND ASSOCIATIONS.

Since Food Inspection Decisions Nos. 76 and 77 were published recommendations of lists of permitted colors have been made by other individuals and by a voluntary association. For the purpose of comparing these proposed lists of permissible colors with the permitted list of Food Inspection Decision No. 76, the former are now to be examined in the same manner as the coal-tar colors, on the United States market in the summer of 1907 for food coloring pur-

poses, were examined in order to establish the permitted list of 7 colors. These recommendations cover the following lists:

- 1. W. Ernst's list.
- 2. Muttelet's interpretation of the French law.
- 3. Second International White Cross Congress list.
- 4. Beythien and Hempel's list.
- 5. An American manufacturer's list.
- 6. Béhal's list.

#### ERNST.

W. Ernst (Färber Zeitung 1908, vol. 19, p. 381; abst. Chem. Ztg. "Reportorium" 1909, p. 89) recommends for use in foods the following 38 titles of coal-tar colors. The Green Table numbers appear in parentheses after each title where corresponding numbers could be ascertained; the italicized numbers are those of the permitted list of Food Inspection Decision No. 76.

- 1. Auramin (425, 426).
- 2. Naphthol Yellow S (4).
- 3. Quinolin Yellow (666, 667).
- 4. Tartrazin (94).
- 5. Acid Yellow (8, 88, 95, 4).
- 6. Spirit Yellow (Amidoazobenzol) (7).
- 7. Curcumin S (399).
- 8. Eosin (512, 514, 515, 517, 521).
- 9. Erythrosin (516, 517).
- 10. Fluorescein (510).
- 11. Orange II (86).
- 12. Crocein Orange (13).
- 13. Basic Oranges (?).
- 14. Fast Red (63, 65, 102, 103, 105, 107, 144).
- 15. Amara Red (?).
- 16. Naphthol Red (?).
- 17. Azo red (62).
- 18. Bordeaux (65, 107, 157, 170, 171, 198, 244).
- 19. Victoria Rubin (?).
- 20. Ponceau (13, 15, 44, 55, 56, 57, 108, 114, 146, 147, 148, 150, 160, 163, 169, 448).

- 21. Rhodamin (496, 497, 498, 502, 504, 505).
- 22. Acid Magenta (462).
- 23. Safranin (583, 584, 585).
- 24. The Croceins (13, 104, 106, 145, 151, 160, 164, 169).
- 25. Acid Green (434, 435).
- 26. Brilliant Fulling Green (?).
- 27. Malachite Green (427, 428).
- 28. Brilliant Green (428).
- 29. Water Blue (480).
- 30. Patent Blue (440, 442).
- 31. Brilliant Fulling Blue (?).
- 32. Domingo Blue B extra (?).
- 33. Methyl Violet (451, 454).
- 34. Acid Violet (464, 465, 467, 468, 470, 471, 472, 474, 507).
- 35. Bismarck Brown (197, 201).
- 36. Acid Brown (133, 138).
- 37. Nigrosin (600,602).
- 38. Several Acid Blacks (184 and ?).

Of these 38 titles only 11 refer to a single entry each in the Green Tables (namely 2, 4, 6, 7, 10, 11, 12, 17, 22, 28, 29); 7 titles can not be definitely connected with any entry in the Green Tables (titles 13 15, 16, 19, 26, 31, 32); one title is broader than the corresponding color in the Green Tables (title 38) and the remaining 19 titles each and all refer to more than one entry in the Green Tables.

These 31 titles embrace 88 different entries in the Green Tables; some of these entries are included in two or more titles as follows:

Green Table number.	Titles.	Green Table number.	Titles.
4	 2,5	160	20, 24
		169	
65	 14,18	428	27, 28
107	 14, 18	517	8,9

These 88 different Green Table entries can be divided as follows on the basis of the compilation of literature on physiological action hereinbefore given (see p. 63):

Unfavorable only.—94, 164, 201, 425, 434, 502, 516, 602, 667. Total 9, or 10.2 per cent.

Favorable only.—4, 65, 102, 103, 105, 107, 169, 399, 435, 462, 467, 512, 517, 521, 600. Total 15, or 17.0 per cent.

Conflicting.—8, 13, 15, 55, 86, 88, 95, 106, 138, 160, 163, 197, 427, 428, 448, 451, 480, 504, 584. Total 19, or 21.6 per cent.

Not reported on.—7, 44, 56 ¹, 57, 62, 63, 104, 108, 114, 133, 144, 145, 146, 147, 148, 150, 151, 157, 170, 171, 184, 198, 244, 426, 440, 442, 454, 464, 465, 468, 470, 471, 472, 474, 496, 497, 498, 505, 507, 510, 514, 515, 583, 585, 666. Total 45, or 51 per cent.

According to this mode of judging only 15, or 17 per cent, of the colors suggested by Ernst for food coloring would be regarded as proper for use in foods.

These 15 embrace 4 of the 7 permitted colors of Food Inspection Decision No. 76, namely, 4, 107, 435, and 517, leaving 11 to be considered. Of these, 5 were not on the United States market in the summer of 1907, namely, 102, 399, 467, 521, and 600.

The remaining 6 are as follows (the numbers in parentheses being the number of sources out of a possible 12 offering them on the United States market in the summer of 1907): 65 (2); 103 (6); 105 (1); 169 (1); 462 (2); and 512 (3).

The reasons for the noninclusion of these in the permitted list of Food Inspection Decision No. 76 have been given on page 167.

## MUTTELET'S INTERPRETATION OF THE FRENCH LAW.

Muttelet (Annales des Falsifications, 1909, pp. 26-38), places the following interpretation on the French regulations of December 29, 1890, and of August 4, 1908, classifying them as—

- I. Those colors which are certainly permitted by those regulations.
- II. Those colors whose permitted or prohibited use is doubtful.
- III. Those colors which are certainly prohibited.

The Green Table numbers contained in each class are classified as follows (the italicized numbers being those of the permitted list of Food Inspection Decision No. 76): (a) Unfavorably reported; (b)

<sup>1</sup> See page 167 for special reasons for including Ponceau 3R in permitted list.

favorably reported; (c) conflictingly reported; (d) not reported in the literature as to their physiological action (see p. 63); in parentheses is given the number of sources out of a possible 12 offering those colors on the United States market in the summer of 1907.

- I. Those colors which are certainly permitted:
  - (a) None. (b) 4 (10); 5 (0); 462 (2); 512 (3); 517 (5); 520 (2); 600 (0); total, 7. (c) 55 (2); 427 (2); 451 (5); 457 (0); total, 4. (d) 158 (0); 518 (2); total, 2.
- II. Those colors whose permitted or prohibited use is doubtful:
  - (a) None. (b) 65 (2); 107 (7); 599 (0); total, 3. (c) 8 (5); 84 (2); 427 (2); 457 (0); 601 (1); total, 4. (d) 452 (2); 456 (0); 513 (0); 514 (0); total, 4.
- III. Those colors which are certainly prohibited:
  - (a) 3 (0); (b) none; (c) 427 (2); (d) none.

It will be noted that 427 appears in all three of Muttelet's classifications. The reason for this is that 427 is or has been marketed in at least three different forms; of these the straight chlorhydrate is permitted, the oxalate and the zinc chlorid double salt are of doubtful admissibility, and the picrate is undoubtedly forbidden.

Also 457 appears in Muttelet's Classes I and II because the "Bleu Lumière" of Muttelet's Class I is indistinguishable from his "Bleu Lumière" of Class II, when using the Green Tables as a guide. Further, this list of Muttelet contains only 3 out of the 7 colors permitted in Food Inspection Decision No. 76, namely, 4 and 517 of his Class I and 107 of his Class II.

An inspection of Muttelet's Class I discloses 13 Green Table entries, of which only 7 have been reported in the literature, as herein compiled and rated, in a favorable manner; of these 7, 2 were not on the United States market in the summer of 1907 and of the remaining 5, 2 are in the permitted list of Food Inspection Decision No. 76; the remaining 3 are: 462. Acid Magenta (2); 512. Eosin (3); 520. Rose Bengal (2), and the reasons for whose noninclusion in the permitted list of Food Inspection Decision No. 76 have been given (p. 167).

## SECOND INTERNATIONAL WHITE CROSS CONGRESS.

The Second International White Cross Congress, held in Paris, October 18 to 24, 1909, according to the Chemiker Zeitung, 1909, page 1227, adopted the following list of colors which were said to be proper for use in coloring food products. The figures in parentheses are the Green Table numbers; the italicized numbers are those of the permitted list of Food Inspection Decision No. 76.

- 1. Erythrosin (516, 517).
- 2. Rhodamin B (504).
- 3. Bordeaux S (107).
- 4. Bordeaux G (170).

- 5. Fast Red E (105).
- 6. New Coccin (106).
- 7. Ponceau 2R (55).
- 8. Xylidin Scarlet (55).

- 9. Magenta (448).
- 10. Acid Magenta (462).
- 11. Orange I (85).
- 12. Naphthol Yellow S (4).
- 13. Chrysoin (84).
- 14. Auramin O (425).
- 15. Acid Green (434, 435).

- 16. Lyons Blue (457).
- 17. Patent Blue (440, 442).
- 18. Paris Violet (451).
- 19. Acid Violet (464, 465, 467, 468, 470, 471, 472, 474, 507).
- 20. Black Indulins (599).
- 21. Sulphonated Nigrosin (602).

It will be observed that titles 7 and 8 refer to the same Green Table number; this leaves, therefore, only 20 titles to consider. These refer to 31 different Green Table numbers which are classified as (a) only unfavorable reports, (b) only favorable reports, (c) conflicting reports, and (d) no reports, in the literature hereinbefore compiled and rated (see p. 63):

- (a) 425, 434, 516, 602; total, 4.
- (b) 4, 85, 105, 107, 435, 462, 467, 517, 599; total, 9.
- (c) 55, 84, 106, 448, 451, 457, 504; total, 7.
- (d) 170, 440, 442, 464, 465, 468, 470, 471, 472, 474, 507; total, 11.

The numbers of section (b), of which only favorable reports are recorded, are the only ones here of interest; they are 9 in number, or less than 30 per cent of all those included in this list, and of these 9, 5 are on the permitted list of Food Inspection Decision No. 76. The remaining 4 are the following, the number in parentheses representing the number of sources out of a possible 12 offering them on the United States market in the summer of 1907:

- 105. Fast Red E (1).
- 462. Acid Magenta (2).
- 467. Acid Violet 6 B, not offered.
- 599. Printing Blue, not offered.

The reasons for the noninclusion of these colors in the permitted list of Food Inspection Decision No. 76 have been given on page 167.

#### BEYTHIEN AND HEMPEL.

Beythien and Hempel (Fürber Ztg., 1909, v. 15, pp. 301, 348, 392, 436; abst. Chem. Ztg., 1910, p. 58) recommend the following colors for use in food products. (The numbers in parentheses are the corresponding Green Table numbers, where such connection could be established; the italicized numbers are those of the permitted list of Food Inspection Decision No. 76.)

- 1. Alizarinblue (562, 563).
- 2. Amaranth (107).
- 3. Bordeaux Red (?).
- 4. Brilliant Blue (?).
- 5. Diamondfuchsin (448).
- 6. Fast Blue (477, 599, 601, 639, 640).
- 7. Fast Yellow R (9).
- 8. Fast Red (63, 65, 102, 103, 105, 107, 144).
- 9. Eosin (512, 514, 515, 517, 521).
- 10. Erythrosin (516, 517).
- 11. Fuchsin S (462).
- 12. Light Green S F yellowish (435).
- 13. Indigo disulphoacid (692).
- 14. Indulins (599, 601, 603).
- 15. Light Blue (?).
- 16. Malachite Green (427, 428).
- 17. Methyl Violet (451, 454).

```
18. Naphthol Yellow S (4).
                                            26. Roccellin (102).
19. Orange I (85).
                                            27. Roscellin (?).
20. Orange L (54).
                                            28. Rubin (448).
21. Paris Violet (451).
                                            29. Acid Yellow S (4).
22. Phloxin (518, 521).
                                            30. Acid Magenta (462).
23. Ponceau 3R (56, 57).
                                            31. Solid Blue (?).
24. Ponceau Red (?).
                                            32. Tropaeolin OOO (85, 86).
25. Primrose (3, 513, 514).
                                           33. Waterblue (480).
```

For six titles (3, 4, 15, 24, 27, and 31) no corresponding Green Table number could be determined.

Of the remaining 27 titles, No. 9 includes part of No. 10, 32 includes 19, 9 includes part of 25, 8 includes 26, 17 includes 21, 11 is identical with 30, and 18 is identical with 29, all on the assumption that the correct connections between title and Green Table numbers have been made. Therefore these 27 titles are, in fact, only 25 titles; of these 25 titles, 13 refer to but one Green Table number each; the remaining 12 titles each refer to two or more Green Table numbers.

The Green Table numbers above given are now arranged in the following four classes according to the literature hereinbefore compiled and rated (see p. 63):

- (a) Only unfavorable reports: 3, 516, 639; total, 3.
- (b) Only favorable reports: 4, 65, 85, 102, 103, 105, 107, 435, 462, 477, 512, 517, 521, 599, 692; total, 15.
  - (c) Conflicting reports: 9, 86, 427, 428, 448, 451, 480, 563, 601; total, 9.
  - (d) No reports: 54, 56, 157, 63, 144, 454, 513, 514, 515, 518, 562, 603, 640; total, 13.

The 15 Green Table numbers of class (b) are the only ones here of interest; they include 6 out of the 7 permitted colors of Food Inspection Decision 76; of the remaining 9, 4 (102, 477, 521, 599) were not on the United States market in the summer of 1907. The remaining 5 are as follows. (The number in parentheses is the number of sources out of a possible 12 offering them on the United States market in the summer of 1907):

```
65. Fast Red B (2). 462. Acid Magenta (2). 103. Azorubin S (6). 512. Eosin (3). 105. Fast Red E (1).
```

The reasons for the noninclusion of these colors in the permitted list of Food Inspection Decision No. 76 have been given on page 167.

## SUMMARY OF THREE PRECEDING RECOMMENDATIONS.

The recommendations made by Ernst, the White Cross Congress, and Beythien and Hempel are summarized in the following table:

<sup>&</sup>lt;sup>1</sup> See page 167 for special reasons for permitting use of Ponceau 3R.

Summary of recommendations from three sources.

G	Unfavo	rable repor	ts only.	Number	
Green Table No. (11).	Ernst.	White Cross.	Beythien and Hempel.	recom- mending.	Dealers offering.
3	x x x x x x x x	x x x	x	1 1 1 1 2 2 2 1 3 2 1	0 6 0 2 3 1 2 1 0 0
Total	9	4	3		

G	Favora	able report	s only.	Number	
Green Table No. (19).	Ernst.	White Cross.	Beythien and Hempel.	recom- mending.	Dealers offering.
4	x x x x x x x x x x x x x	x x x x x x	x x x x x x x x x x x x x x x x x x x	3 2 2 2 2 3 3 1 1 3 3 2 1 2 3 2 2 1 1	10 2 2 0 6 1 7 1 0 4 2 0 0 3 5 0 0 3
Total	15	9	15		

Green	Con	flicting rep	orts.	Number	
Table No. (24).	Ernst.	White Cross.	Beythien and Hempel.	recom- mending.	Dealers offering.
8 9	x		x	1 1	5 1
13 15 55	x x x	х		$egin{array}{c} 1 \ 1 \ 2 \end{array}$	6 0 2
84 86 88	x	x	х	1 2 1 1 2 1 2	0 2 2 8 0 2 5
95 106	x x x	x		$\frac{1}{2}$	2 5
138 160 163	X X X			1	0 0 0
197 427 428.	X X X		x x	1 2	0 4 2 3 4 5
448	X X	X X	X X	3 3	5 5
457 480 504	x x	x x	x	1 1 2 2 3 3 1 2 2	0 1 5
563 584	х		x	1 1 1	0 1 1
Total	19	7	9		

Summary of recommendations from three sources—Continued.

Green Table No. (51).	Ernst.	White Cross.	Beythien	Number recom-	Dealers
			and Hempel.	mending.	offering.
444 554 56 57 62 63 104 108 114 133 144 145 146 117 118 150 151 157 170 171 188 244 440 442 455 468 470 471 472 474 496 497 498 505 507 510 513 514 555 518	X X X X X X X X X X X X X X X X X X X	x x x x x x x x x x x x x x x x x x x	X X X X X X X X X X X X X X X X X X X	1112211111211112212222222211112211	0 0 1 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
583 586 603	x x		x	1 1 1	0
640	x		X	1 1	ŏ
Total.	45	11	13		

These three recommendations embrace 105 different Green Table numbers. The following table discloses the conformity of the composite of these three recommendations to the United States market in the summer of 1907:

Number of suggested samples in the three supplementary lists found on the market, 1907.

Data.	Total in class.	On United States market.
Unfavorable. Favorable. Conflicting. Not reported. Total.	19 24	7 13 17 10 47

## UNPUBLISHED RECOMMENDATIONS OF A MANUFACTURER.

In addition to these published recommended lists the following recommendation has been made by letter by a manufacturer who contributed specimens of food colors to the United States market in the summer of 1907:

That for the following permitted colors of Food Inspection Decision No. 76, there be substituted certain colors, the Green Table numbers alone being here given:

Permitted.	Substitute.
4	94
56	53
85	- 15
517	521

In regard to these proposed substitutes it is to be said that Nos. 15 and 521 were not on the United States market in the summer of 1907, according to the canvass made and described in Section I, Nos. 53 and 94 were on that market, No. 53 had not been examined physiologically, and No. 94 had been examined physiologically with only unfavorable results. The way was, therefore, not open to placing any of these colors on the permitted list under the procedure adopted. However, had No. 15 been on the market it would not have been placed on the permitted list, because it has been examined physiologically with contradictory results, while No. 521 might have been placed on the permitted list because it seems to have been examined physiologically and with only favorable results.

#### BÉHAL.

As the result of careful investigation Béhal (Revue Générale des Matières Colorantes, 1910, p. 131) suggests the use of 21 definite chemical individuals which are given in the following. With the tabulation on page 63 as a guide these 21 colors are classified as unfavorable, favorable, contradictory, and not reported on. The italicized figures are in the permitted list of Food Inspection Decision No. 76.

UNFAVORABLE (1).

425. Auramin O (3).

#### FAVORABLE (11).

```
      4. Naphthol Yellow S (10).
      462. Acid Magenta (2).

      65. Bordeaux B (2).
      467. Acid Violet 6B (none).

      85. Orange I (2).
      512. Eosin (2).

      105. Fast Red (1).
      517. Erythrosin (5).

      107. Amaranth (7).
      520. Rose Bengal (2).

      435. Light Green S F yellowish (4).
```

97291°-Bull, 147-12-12

### CONTRADICTORY (6).

55.	Ponceau 2 R (2).	427. Malachite Green (2).
84.	Chrysoin (2).	427. Malachite Green (2). 451. Methyl Violet (5). 480. Water Blue (1).
106.	New Coccin (5).	480. Water Blue (1).

#### NOT REPORTED ON (3).

```
54. Scarlet R (1).
64. Crystal Ponceau (1).
```

The five in the "favorable" list which were on the United States market in the summer of 1907, but are not in the permitted list of Food Inspection Decision No. 76, namely:

```
65. Bordeaux B (2);
105. Fast Red (1);
462. Acid Magenta (2);

512. Eosin (2);
520. Rose Bengal (2),
```

are tinctorially provided for in that list; only one of the 21 colors recommended by Béhal was not on the market of the United States in the summer of 1907, namely, 467 Acid Violet 6B, and this is in the "favorable" list. The reasons for the non-inclusion of these colors are given on page 167.

#### CONCLUSIONS.

It is clear from an inspection of the preceding analysis of the six lists just given that the confusion referred to on page 165 as existing in the three recommendations made before the formulation of the permitted list of Food Inspection Decision 76 is not at all diminished by these six lists published after the announcement of that decision, as is shown in the following table:

Analysis of six lists published after the issuance of F. I. D. 76.

	Total of	Report	s on physi effect are—	ological -	Physio- logical	Number of dyes wanted
List of—	dyes wanted.	Unfavor- able.	Favor- able.	Contra- dictory.	effect not re- ported.	and contained in F. I. D. 76.
Ernst. Muttelet's class I. White Cross. Beythien and Hempel Manufacturer Béhal	88 7 31 40 4 21	9 4 3 1 1	15 7 9 15 1	7 9 1 6	45 11 13 1 3	4 2 5 6 0 5

The same wide differences of opinion as to the four points discussed on page 166 not only continue but are accentuated.

# XIV. CHEMICAL EXAMINATION OF THE SEVEN PERMITTED COLORS, 1907.

### NEED OF CHEMICAL CONTROL.

The most striking thing, from the chemist's point of view, in the literature relative to the physiological action of coal-tar colors, is the almost universal absence of the results of chemical examination or identification of the materials subjected to physiological test. There is hardly any description or statement of the strength or the concentration of the materials examined, or of the amount or nature of materials not coloring matter present in the substances subjected to these physiological tests. It is true that some publications give identification tests, but few assert the identity of the material subjected to certain physiological tests, with the description so given.

The need for some such chemical control as to identity and quality must be apparent to all having experience with the commercial varieties or grades of coal-tar colors. Many of these commercial brands of coal-tar colors contain added coloring matter other than the principal constituent for the purpose of correcting some defect unavoidably arising during the manufacture. Where those commercial brands are sold for textile and other manufacturing purposes there is no valid objection to such practice, because in such uses the tinctorial properties and effects of the commercial brands are the things the buyers desire and pay for, and the exact nature of the materials accomplishing the results is in reality of secondary importance; but when coal-tar colors are to be used in foods, and in addition to tinctorial effect the absence of any physiological action is necessary, it is at least of doubtful propriety to market such corrected or adjusted brands unless the correction or the adjustment be made with harmless colors.

There are a few instances in the relevant literature where it was attempted to explain the different results attained by observers by the difference in the composition of the materials subjected to physiological test, and there are other instances showing the difficulties encountered in obtaining specimens of coal-tar colors which would always give favorable physiological result.

The variation in composition of coal-tar colors may be exemplified

by the following excerpts from the literature:

1. Weyl (p. 91), discussing the differences between his results and those of Cazeneuve and Lépine with Naphthol Yellow S, states as follows:

It is to be noticed, however, that Cazeneuve and Lépine state that the Naphthol Yellow used (Jaune NS) was difficultly soluble, so that they probably experimented

with another substance. My own investigations were made on dogs with a preparation for which I am indebted to the kindness of Dr. G. Schultz, of the Aniline Manufacturing Co. of Berlin. It was purified by precipitation and recrystallization.

- 2. Fraenkel (p. 578) expresses the opinion that the harmful results observed with Metanil Yellow (95 of the Green Tables) may be due to diphenylamin contained in the color, which diphenylamin may be a decomposition product of the Metanil Yellow, particularly since, as Weyl states (p. 130), it "smelled strongly of diphenylamin."
- 3. Pfeffer (Unterschungen aus dem Botanischen Institut zu Tuebingen, Vol. II, p. 186) says:

In repeating my experiments I beg that it be considered that differences in observation may be caused by the quality of the coloring matters. For quite apart from the fact that the coloring matters are in part variable mixtures of various compounds, and not infrequently contain foreign admixtures, the quality of the goods brought into market may change from time to time. Thus formerly magenta was the acetate, whereas to-day it is the hydrochlorid. Also foreign admixtures may be poisonous or may exert no influence on the absorption of color.

4. Stilling (Archiv. Exper. Path. Pharm., vol. 28, 1891, p. 352) says:

With respect to the blue pyoctanin, I have during the entire time been engaged in ascertaining the most effective. The things marketed by E. Merck as P. cœruleum have therefore now become uniform and homogeneous, which could not be the case at the beginning. The P. cœruleum now furnished by E. Merck is the hydrochlorid of pure hexamethylpararosanilin.

The variable degrees of purity attained by products of this kind on the market was apparently the first cause of the prohibition by the German Government of the coal-tar color known as Corallin for use in food products—not because Corallin itself was harmful, but because in the manufacture of this product it was so apt to retain impurities which in and of themselves produced bad effects, so that this particular color was specifically excluded by the German Government.

In a few cases it is stated that the substance subjected to physiological test had been purified, but these descriptions are hardly such as to enable others to arrive at substantially the same result with reasonable certainty.

5. Stilling in his monograph entitled Anilin Farbstoffe als Antiseptica (Strassburg, 1890, Pt. I, p. 16) says:

The foregoing (relative to the action of Methyl Violet on the eyes of rabbits) holds only for pure substances. Many anilin colors of otherwise very antiseptic properties are contaminated with arsenic, particularly the otherwise useful Ethyl Violet. With such substance serious poisonings and death can be produced in experimental animals.

6. Stilling in Part II of the same monograph, pages 5 and 6, says:

Such substances (coal-tar colors) must be chemically pure and can not, for example, be any mixture of various blue or violet dyes. There is therefore a great difference whether a substance be tested only bacteriologically or also in addition physiologically and therapeutically. Two preparations may be antiseptically wholly equal, but the one preparation may contain harmful admixtures which produce violent irritations.

7. Thus Weyl, speaking of his Metanitrazotin (p. 120), describes the purification method as follows:

It is dissolved in warm alcoholic solution of sodium hydroxid filtered and precipitated with hydrochloric acid. The precipitate is freed from the adhering liquid by the aspirator and washed with hot water.

8. Weyl says of his Metanil Yellow (p. 130):

For purification the color was dissolved in water filtered and separated by the addition of sodium acetate. The yellow mass was freed from the adherent liquid by the filter pump and dissolved in hot alcohol, in which it is difficultly soluble, and obtained from this in the form of yellow crystals. The material used for the experiment was almost pure, as the following analytical statement shows: 0.4895 gram of the color dried in 105° gave 0.084 sodium sulphate. Sodium required, 6.1; found, 5.6. (Note.—This amounts to 91.8 per cent of theory.)

- 9. Weyl (Zts. Hygiene, 1889, Vol. II, p. 34, On Safranin Poisoning) describes the difficulties he had in obtaining safranin on the market that was clean or pure; all preparations were free from arsenic, and contained small amounts of iron, chlorin, and traces of chromium. In one specimen the ash amounted to 4.8 per cent. The theoretical percentage of nitrogen in pure safranin is, according to Weyl, 15.3 per cent; in two commercial products he found 12.7 and 12.3 per cent, respectively (83 and 80.4 per cent of theory); he recrystallized the specimen containing 12.7 per cent twice from dilute hydrochloric acid; the first recrystallization produced a specimen containing 13.8 per cent (90.2 per cent of theory) of nitrogen; the second recrystallization produced a substance containing 14 per cent (91.5 per cent of theory) nitrogen.
  - 10. Chlopin in his book (p. 110) says:

Nevertheless, each dye was tested by me personally with the usual reagents and the dyeing of fabric in order to avoid the confounding of one dye with another. I convinced myself, from my experience, that not only druggists but the home offices of the makers occasionally send dyes which do not correspond to the requirements and resemble them only in name.

(Page 114.) All the dyes, which according to my experiments proved to be poisonous, were carefully examined for contents of arsenic, chromium, and injurious

metals, and were found to be free from these admixtures.

#### FIRST METHODS OF ANALYSIS USED.

In the summer of 1907 there were on the United States market 30 different specimens of the seven permitted colors of Food Inspection Decision No. 76. A chemical examination of those 30 specimens would disclose the qualitative conditions of the market so far as these seven permitted colors were concerned, and it was expected that certain limits for standards of cleanliness would be fixed by such an examination.

In examining these 30 specimens of the seven permitted colors chemically the following determinations were made:

- 1. Moisture.
- 2. Chlorin as chlorids.
- 3. Sulphated ash, together with its iron, aluminum, calcium, and copper content, and the determination of the sulphuric acid in the sulphated ash.
  - 4. Total sulphur.
  - 5. Gutzeit test (test 17 of the United States Pharmacopæia, eighth revision).
  - 6. Heavy metals test (test 121 of the United States Pharmacopæia).
- 7. Total insolubles, together with the determination of the proportion that is volatile on ignition.
  - 8. Ether extractive.

The methods of analysis actually used on these 30 dye specimens are here given solely for the purpose of comparing them with the methods developed therefrom and presented beginning with page 210. Experience has shown that the methods here given are defective in many particulars, and therefore they are not to be used for exact work.

#### MOISTURE.

Dry a sample of each color weighing 3 grams at  $105^{\circ}$  to  $108^{\circ}$  C. for two hours. The loss in weight is assumed to be moisture.

This method is not wholly accurate in the case of Naphthol Yellow, nor is it accurate in the case of Amaranth; but the scarcity of material made it seem unwise, at this stage, to undertake any extended investigation as to the amount or nature of the heating required surely to expel all moisture. The results, therefore, while not as accurate as might be desired, are, for the purposes of this exploratory investigation, sufficiently accurate for the object for which they were undertaken.

#### CHLORIN AS CHLORIDS.

Gently heat samples weighing 0.1 gram with 2 grams of sodium carbonate, and after destroying the greater part of the organic matter add 0.1 gram of powdered potassium nitrate and gently heat the whole until the organic matter is entirely destroyed. After cooling treat the whole with small amounts of cold water and remove from the crucible; effect the solution of the whole by gently heating. After cooling bring the bulk to about 150 cc, cool to room temperature, slightly acidify with nitric acid, precipitate the chlorin with silver nitrate, and weigh as silver chlorid.

#### SULPHATED ASH.

Moisten half-gram samples with concentrated sulphuric acid, gently evaporate to dryness, and treat the residue with 5 cc of concentrated sulphuric acid and again evaporate to dryness; repeat the operation until a white ash results, when the whole is ignited to constant weight. After weighing take up the sulphated ash in boiling water (and if necessary, any undissolved material can be taken up with hydrochloric acid by treatment on a boiling-water bath.) Mix the two solutions and bring to a total volume of 200 cc. In one half thereof determine the contained sulphur by a precipitation with barium chlorid, and weigh as barium sulphate; in the other half determine iron and aluminum by precipitation with ammonia, ignite and weigh as ferric and aluminum oxid. No separation of any contained aluminum was under-

taken in the filtrate from the iron; the lime was precipitated as oxalate. In the only case that copper was encountered it was precipitated as sulphid before taking out the iron or the calcium.

#### TOTAL SULPHUR.

Mix 0.2 gram of the sample with 4 grams of sodium carbonate and 0.5 gram of potassium nitrate, and ignite to complete the destruction of the organic matter; take up in hot water; acidify with hydrochloric acid and precipitate the sulphur as barium sulphate with barium chlorid.

#### GUTZEIT TEST.

Mix 2 grams of the substance with 2 grams of a mixture of 1 part of potassium nitrate and 5 parts of sodium carbonate and ash in a porcelain crucible over a low flame; if not white when cool, mix the ash with 1 gram of potassium nitrate and again ash over a low flame. Generally the second ashing is sufficient. Dissolve the residue in 50 cc of hot water, boil, filter, neutralize with dilute sulphuric acid, and evaporate to substantial dryness on a boiling-water bath. Then add 1 cc of concentrated sulphuric acid and dry over a Bunsen flame; take up the residue with 5 cc of distilled water containing 0.5 cc of concentrated sulphuric acid and 10 cc of a saturated solution of sulphurous acid; evaporate the whole to a bulk of 5 cc on a water bath; add 20 cc of 8 per cent hydrochloric acid and subject this material to the action of 2 grams of metallic zinc, free from arsenic, which has been so activized by means of platinic chlorid that at the end of two hours more than 1 gram of zinc has dissolved and the evolution of gas has been constant and continuous. Conduct the reaction in a flask of 60 cc capacity with a neck 1 cm in diameter and 6 cm long. After the introduction of the solution and the zinc into the flask, stopper the neck of the bottle with gauze, the lower half of which is dry and the upper half moistened with the test solution of lead acetate of the United States Pharmacopæia. After carefully wiping the lip of the flask, cover it with Schleicher & Schüll quantitative filter paper which has been saturated three times with alcoholic mercuric chlorid solution, with complete drying between each saturation on one and the same spot of the filter paper.

In the case of Naphthol Yellow S it is necessary to heat gently with 10 grams of sodium carbonate until the organic matter is substantially all destroyed, then add 1.5 grams of potassium nitrate and heat to complete destruction of the organic matter. Dissolve the fused mass in hot water and a few drops of fifth-normal sulphuric acid; then make distinctly acid with the same and add an excess of 1 cc of concentrated sulphuric acid; evaporate first on the water bath and afterwards on asbestos until all odor of nitrous fumes and of hydrochloric acid has disappeared. Take up the residue in 5 cc of water and 15 cc of a saturated solution of sulphurous acid in water. Heat the whole on the water bath until no odor of sulphur dioxid remains.

The preparation of the sample by ignition with carbonate and nitrate should not be done in platinum, but should be done in a porcelain crucible, since it has happened that as much as 0.05 mg of arsenic mixed with the dye, which had been fused in platinum with carbonate and nitrate, could not be detected on the mercury-chlorid paper, whereas 0.01 mg when similarly treated in porcelain could always be detected, and 0.005 mg would usually be found when done in porcelain.

#### HEAVY METALS.

Mix as much of the substance as approximately contains 1 gram of color with 10 times its weight of carbonate of soda and ignite with the addition of 0.5 gram of potassium nitrate. Dissolve the whole in water, any undissolved material being taken up

with hydrochloric acid, bring the two solutions together and slightly acidify with hydrochloric acid; dilute to 100 cc; place 10 cc in a test tube of 40 cc capacity and warm to 50° C. in a water bath; add 10 cc of a freshly prepared saturated solution of hydrogen sulphid in water, stopper the test tube well and allow the whole to stand in water having a temperature of 35° C. for a half hour. Dilute a 3.3 cc portion to 10 cc, treat as before with 10 cc of hydrogen sulphid in water for one-half hour at 35° C., and add to each ammonium hydrate.

#### TOTAL INSOLUBLES.

Dissolve 1 gram of the substance in 1 liter of water, filter through counterpoised quantitative filters, and wash with hot water until all traces of color are removed from the filter paper, then dry at 100° C. to constant weight and weigh; report the weight as total insolubles. Ignite the total insolubles in a platinum crucible and report the residue remaining as nonvolatile insolubles.

#### ETHER EXTRACTIVE.

Two methods were employed, the one consisting in direct extraction of the substance in a Soxhlet apparatus by means of redistilled ether dried over sodium. Schleicher & Schüll extraction cartridges were used after they had been thoroughly extracted by ether and shown by examination that they yielded no extractive to ether whatever. This is a necessary precaution, because the amount of ether extractive matter in these cartridges varies. The amount of ether extractive was determined by driving off the ether over a 32-candlepower incandescent lamp, finally drying and cooling in a desiccator.

This method, however, is not satisfactory, since the results it gives are undoubtedly low and it seems that the higher the material was dried the more erratic were the results. Therefore the following method was used:

Disolve 1 gram of the sample in 100 cc of water; add 0.5 gram of sodium acetate; extract three times with 50 cc of ether in a separatory funnel; mix the ether so recovered with 10 cc of water; separate the ether and dry with fused calcium chlorid; let stand from 12 to 24 hours; pour off from the calcium chlorid; distil off the ether as in the preceding method; acidify the color solution containing sodium acetate with 1 cc concentrated hydrochloric acid; extract three times with 50 cc of ether, and proceed as before.

## RESULTS OF CHEMICAL EXAMINATION, 1907.

## DETAILED CHEMICAL DATA ON EACH PERMITTED COLOR.

The results of this examination are given in the following tabulations under the respective Green Table numbers; the serial numbers refer to the numbers assigned to the specimens as they were received. The abbreviations "p." and "n. p." opposite "Gutzeit test," and "Heavy metals test" stand for "pass," and "not pass," respectively, according as the specimen did or did not comply with those requirements; the entries opposite "Ratio," indicate the degree of concordance of the sulphated ash as weighed, and the contained  $SO_3$  calculated back to sodium sulphate and is a measure of the accuracy or dependability of the sulphated ash item and its determination.

As a guide in ascertaining the conformity of these colors to their supposed standards when judged by the analytical data obtained, the following table may of service:

Percentages of sulphur, sodium, and sulphated ash properly belonging to each of the seven permitted colors.

Green Table No.	Name of colors.	Sulphur.	Sodium.	Sul- phated ash.
4 56 85 107 435 517 692	Naphthol Yellow. Ponceau 3 R. Orange I Amaranth. Light Green. Erythrosin. Indigotin.	12. 97 9. 16 15. 92 11. 12	12. 87 9. 33 6. 58 11. 44 8. 31 5. 24 9. 89	39. 73 28. 80 20. 31 35. 32 25. 64 16. 17 30. 52

1 Iodin in No. 517=57.7 per cent.

Detailed analytical data obtained on several samples of each of the permitted colors (per cent).

GREEN TABLE NO. 4. NAPHTHOL YELLOW S.

		1	Serial Nos.		
Determinations.	23.	73.	108.	142.	187.
Moisture NaCl Na as NaCl Sulphated ash: Al and Fe	0. 70 6. 73 2. 65	0. 28 3. 47 1. 37	0. 28 1. 02 . 40	0. 60 . 61 . 24	0. 33 2. 86 1. 13
Ca Na <sub>2</sub> SO <sub>4</sub> . SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> =Na <sub>2</sub> SO <sub>4</sub> . Ratio. Na as Na <sub>2</sub> So <sub>4</sub> . Total sulphur.	.04 38.00 38.18 100.3 12.32 8.72	None. 48. 8 49. 57 101. 6 15. 82 9. 47	None. 47. 1 47. 32 100. 5 15. 46 12. 63	None. 51. 4 51. 76 100. 7 16. 66 12. 42	None. 49. 2 49. 44 100. 5 15. 95 12. 32
Gutzeit test Heavy metals test. Total insolubles Nonvolatile. Volatile Acetate ether extract Acid ether extract. Total ether extract.	n.p. n.p. .05 .03 .02 .34 .98	n.p. n.p. None. None. None. . 15 . 90 1.05	n.p. n.p. None. None. None. . 14 . 70	n.p. n.p. None. None. . 16 . 75 . 91	n.p. n.p. . 05 None. . 05 . 10 . 32 . 42
Ether extract solid	.062	.042	.04	.03	.022
	1				
Determinations			Serial Nos.		
Determinations.	201,	209.	Serial Nos.	272.	280.
Moisture	201. 0.50 23.67 9.31	<u> </u>	1		280. 9.07 9.39 3.70
Moisture NaCl. Na S NaCl. Sulphated ash: Al and Fe Ca NagSO <sub>4</sub> . Sog in Na <sub>2</sub> SO <sub>4</sub> = Na <sub>2</sub> SO <sub>4</sub> . Ratio. Na as Na <sub>2</sub> SO <sub>4</sub> .	0.50 23.67 9.31 .21 .03 49.20 49.75 101.1 15.95	209. 0. 60 4. 08 1. 61 None. None. 53. 10 53. 76 101. 3 17. 22	228. 0. 32 2. 86 1. 13 .07 None. 55. 60 56. 45 101. 5 18. 03	2.12 3.26 1.28 .07 None. 45.60 45.80 100.4 14.79	9. 07 9. 39 3. 70 .07 .07 40. 20 40. 14 99. 84 13. 03
Moisture Na Cl Na as NaCl Sulphated ash: Al and Fe Ca NagSO4 SO3 in NagSO4= NagSO4	0.50 23.67 9.31 .03 .49.20 49.75 101.1	209. 0. 60 4. 08 1. 61 None. None. 53. 10 53. 76 101. 3	0. 32 2. 86 1. 13 . 07 None. 55. 60 56. 45	2.12 3.26 1.28 .07 None. 45.60 45.80 100.4	9. 07 9. 39 3. 70 .07 .07 40. 20 40. 14 99. 84

## GREEN TABLE NO. 56. PONCEAU 3R.

Determinations.	Serial No. 9.	Determinations.	Serial No. 9.
Moisture NaCl Na as NaCl Sulphated ash:	7.84 .28 None. 38.10	Na as Na <sub>2</sub> SO <sub>4</sub>	12.36 p. n.p. .28 .83 1.11

## GREEN TABLE No. 85. ORANGE I.

Determination	Serial Nos.		Determinations	Serial Nos.	
Determinations.	16.	224.	Determinations.	16.	224.
Moisture. NaCl. Na as NaCl. Sulphate ash: Al and Fe Ca. Na <sub>2</sub> SO <sub>4</sub> . SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> = Na <sub>2</sub> SO <sub>4</sub> . Na as Na <sub>2</sub> SO <sub>4</sub> .	11. 06 8. 78 3. 18 .14 None. 30. 80 21. 18 101. 3 9. 99	5. 39 3. 51 1. 38 .07 .14 18. 00 18. 09 100. 50 5. 84	Total sulphur. Gutzeit test. Heavy metals test. Total insolubles. Nonvolatile. Volatile. Acetate ether extract Acid ether extract Total ether extract. Ether extract solid.	10. 12 p. p. 1. 50 . 20 1. 30 . 02 . 20 . 22 . 214	8. 34 n.p. p. . 55 . 20 . 35 . 62 . 20

## GREEN TABLE No. 435. LIGHT GREEN SF YELLOWISH.

		Serial	Nos.	
Determinations.	57.	92.	168.	233.
Moisture. NaCl. Na as NaCl. Sulphate ash: Al and Fe. Ca. Na <sub>2</sub> SO <sub>4</sub> . SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> = Na <sub>2</sub> SO <sub>4</sub> . Ratio. Na as Na <sub>2</sub> SO <sub>4</sub> . Total sulphur Gutzeit test Heavy metals test. Total insolubles Nonvolatile Volatile Acetale ether extract. Acid ether extract.	3. 29 9. 00 7. 78 86. 46 2. 92 11. 48 Tr. (p.) p. . 20 None. . 20 . 02 . 05	5. 08 . 102 . 040 . 28 . 00 43. 40 43. 96 101. 30 14. 07 15. 44 Tr. (p.) p. . 45 . 69 . 39 . 02 . 05 . 07	5. 04 1. 735 . 683 . 14 . 085 48. 00 46. 68 95. 92 15. 82 Heavy. p. . 75 . 60 . 15 . 05	4. 15 1. 530 . 603 . 14 . 0.85 53. 00 53. 62 101. 3 17. 19 16. 75 Heavy. P. . 95 . 60 . 35 None. . 05

## GREEN TABLE No. 107. AMARANTH.

Determinations.	Serial Nos.								
Determinations.	82.	96.	130.	162.	177.	219.			
Moisture	4. 24	6. 46	6. 23	9. 81	4. 20	8. 24			
NaCl.	38. 91	12. 79	35. 79	25. 39	28. 45	24. 21			
Na as NaCl.	15. 32	5. 04	14. 10	10. 00	11. 21	9. 54			
Sulphate ash: Al and Fe. Ca. Na <sub>2</sub> SO <sub>4</sub> . SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> =Na <sub>2</sub> SO <sub>4</sub> .	.14	. 07	.14	. 42	. 42	. 21			
	.215	. 857	.93	. 428	. 428	. 57			
	65.20	53. 20	59.80	48. 80	60. 00	49. 80			
	66.50	55. 60	60.30	49. 52	61. 26	50. 72			

Detailed analytical data obtained on several samples of each of the permitted colors (per cent)—Continued.

GREEN TABLE No. 107. AMARANTH-Continued.

	Serial Nos.							
Determinations.	82.	96.	130.	162.	177.	219.		
Ratio Na as Na <sub>2</sub> SO <sub>4</sub> . Total suiphur Copper Gutzeit test. Heavy metals test. Total insolubles Nonvolatile Volatile Acetate ether extract. Acid ether extract. Total ether contract. Ether extract solid	102.00 21.13 8.62 None. n. p. n. p. .30 .12 .18 .08 .07 .15	100. 80 17. 22 13. 02 None. n. p. n. p. 1. 50 1. 10 . 40 . 15 . 15 . 30 . 062	100. 80 19. 39 8. 57 None. n. p. 2. 05 1. 10 . 95 . 18 . 15 . 33 . 052	101. 5 15. 82 9. 42 .09 n. p. n. p. 1. 05 .70 .20 .03 .23 .040	102.1 19.45 11.09 None. n. p. .90 .35 .55 .02 .03 .05	101. 9 16. 14 8. 54 None. n. p. n. p. 2. 40 1. 15 1. 25 . 30 . 10 . 40		

#### GREEN TABLE NO. 517. ERYTHROSIN.

	Serial Nos.				
Determinations.	184.	200.	216.	254.	
Moisture NaCl (not determined).			9.6		
Sulphated ash: Al and Fe Ca NasSO4.	0.14 None. 39.00	0.14 None. 35.69	.14 None. 31.80	0. 28 No ne 26. 4	
SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> =Na <sub>2</sub> SO <sub>4</sub> . Na as Na <sub>2</sub> SO <sub>4</sub> . Gutzeit test. Heavy metals test.	12.65 p.	35. 44 11. 52 n. p. p.	32. 46 10. 40 p. p.	26. 8 8. 56 p. p.	
Total insolubles. Nonvolatile. Volatile. Acetate ether extract.	. 04 . 56 . 80	None. .20 .35	. 65 . 20 . 45 . 10	. 90 . 20 . 70 . 10	
Total ether extract. Ether extract solid.		. 07	.02	. 02	

### GREEN TABLE NO. 692. INDIGO DISULPHO ACID.

Determine	Serial Nos.					D. d	S	erial No	3.
Determinations.	90.	195.	249.	Determinations.	90.	195.	249.		
Moisture NaCl Na as NaCl. Sulphated ash: Al and Fe Ca Na <sub>2</sub> SO <sub>4</sub> SO <sub>3</sub> in Na <sub>2</sub> SO <sub>4</sub> =Na <sub>2</sub> SO <sub>4</sub> Ratio Na as Na <sub>2</sub> SO <sub>4</sub>	7. 25 7. 02 2. 77 42 . 57 26. 20 26. 74 102. 10 8. 50	5. 32 28. 66 11. 29 .28 .14 55. 40 56. 64 102. 20 17. 97	7. 31 7. 02 2. 77 . 56 . 71 26. 60 27. 36 102. 8 8. 63	Total sulphur Guczeit test Heavy metals test. Total insolubles. Nonvolatile Volatile Acetate ether extract. Acid ether extract. Total ether extract. Ether extract solid.	10.30 p. p.(?) 1.55 .55 1.05 .02 .05 .07 .002	10.30 p. p (?) .80 None. .80 .02 .10 .12 .014	10. 00 pp (? 1. 14 . 56 . 66 . 06 . 06 . 06 . 06 . 06 . 06		

# RECALCULATION OF ANALYTICAL DATA ON BASIS OF COLORING MATTER PRESENT.

The difficulties in the way of translating these analytical data into proximate constituents are so great, in so many of the cases, as to make any attempts to obtain practical results in that way absolutely

useless; the amount of material on hand was unfortunately so small at the beginning (2 ounces or less, in most instances) that the utmost economy of material was necessary to get the data reported, on account of the large amount of material needed in the exploratory work done in trying out the methods for determining arsenic, heavy metals, and ether extractives.

In the arsenic test the coloring matter, as a whole, was considered, and no attempt was made to get at the actual amount of real coloring matter in the exact weights of the material as a whole, used for such examination, and consequently the results are not translatable into actual weights of real color used. It is obvious that if 2 grams, or other weight of the substance as a whole, failed to pass any test when the whole amount was considered as though it were all color, the material could not possibly have passed those tests if amounts thereof, corresponding to the prescribed weights of actual color used, were taken; also it is clear that if the sample, as a whole, passed a given test, at a given weight thereof, it would not necessarily have passed that test had an amount thereof, corresponding to that same weight of actual color, been used.

The amount of actual or real coloring matter in 27 of these 30 specimens is not greater than shown in the following:

Per	$cent\ of$	actual	coloring	matter	in	27	samples.
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Name of color.	Serial No.	Per cent.	Serial No.	Per cent.
Naphthol Yellow S.	23 73 108 142	91.20 95.20 97.86 97.88	201 209 228 272	75.56 93.19 95.72 92.81
Ponceau 3R Orange I Amaranth	. 16	96. 34 73. 24 78. 44 56. 40 78. 95	280 224 162 177	79. 47 89. 76 63. 52 66. 40
Light Green SF Yellowish  Erythrosin Indigo Disulpho Acid	09	55.60 91.37 94.30 89.65 84.11	219 168 233	64. 25 92. 37 93. 32

In order to make these analytical results comparable among themselves and with other analytical results later to be given, the items common salt, volatile and nonvolatile insolubles, total insolubles, acetate ether extract, acid ether extract, and total ether extract of the preceding tabulations have been recalculated in parts per 100 of actual or real coloring matter as just enumerated:

## Analytical data recalculated to parts per hundred of coloring matter present.

## GREEN TABLE NO. 4. NAPHTHOL YELLOW S.

To de contractivo	Serial Nos.									
Determinations.	23.	73.	108.	142.	187.	201.	209.	228.	272.	280.
Common salt	7.38	3.64	1.04	0.62	2.97	31. 53	4.38	2.99	3.51	11.82
Insolubles: Volatile. Nonvolatile.	.02	.00	.00	.00	.05	.13	.86	.02	.97	1.40
Total	.05	-00	.00	.00	.05	.40	.86	.12	.97	1.5
Ether extract: Acetate. Acid.	.37	.16	.14	.16	.10	.19	.27 1.16	.08	.16	.13
Total	1.44	1.11	.86	. 93	.43	.63	1.43	1.02	. 94	1.10

## GREEN TABLE NO. 56. PONCEAU 3R.

Determinations.	Serial No. 9.	Determinations.	Serial No. 9.
Common salt		Ether extract: Acetate. Acid	0.38 1.13
Nonvolatile		Total	1.51

## GREEN TABLE NO. 85. ORANGE I.

Determinations	Seria	l Nos.	Determinations.	Serial	Nos.
Determinations.	16.	224.	Determinations.	16.	224.
Common salt	11.19	3.91	Ether extract: AcetateAcid	0.02	0.69
Volatile Nonvolatile	1.66 .25	.39	Total	.27	.92
Total	1.91	.62			

## GREEN TABLE NO. 107. AMARANTH.

	Serial Nos.								
Determinations.	82.	96.	130.	162.	177.	219.			
Common salt	68.89	16.20	64.37	39.97	42.85	37.39			
Insolubles: Volatile Nonvolatile	.32	.51 1.39	1.71 1.98	1.10 .55	.83 .54	1.93 1.78			
Total	.53	1.90	3.69	1.65	1.37	3.71			
Ether extract: Acetate. Acid.	.14	.19	.32	.31	.03	. 46			
Total	.26	.38	.59	.36	.08	.61			

Analytical data recalculated to parts per hundred of coloring matter present—Continued.

GREEN TABLE NO. 435. LIGHT GREEN SF YELLOWISH.

	Serial Nos.					
Determinations.	57.	92.	168.	232.		
Common salt.	0.04	0.10	1.88	1.64		
Insoluble: Volatile Nonvolatile	.22	.41	.16 .65	.38		
Total	.22	.47	.81	1.02		
Ether extract: Acetate. Acid.	.02	.02	.05	.05		
Total	.07	.07	.10	.10		

#### GREEN TABLE NO. 517. ERYTHROSIN.

Determinations.	Serial No. 216.	Determinations.	Serial No. 216.
Common salt Insoluble: Volatile Nonvolatile Total	0.50	Ether extract: Acetate. Acid. Total.	0.11

#### GREEN TABLE NO. 692. INDIGO SULPHO ACID.

Determinations.	S	Serial No	s.	Determinations.	Serial Nos.			
Determinations.	90.	195.	249.	Determinations.	90.	195.	249.	
Common salt Insoluble:	8.35	44.02	8.31	Ether extract: Acetate. Acid	0.02 .06	0.03 .15	0.02	
Volatile Nonvolatile	1.25 .59	1.23	.77	Total	.08	.18	.06	
Total	1.84	1.23	1.36					

## MARKET QUALITY OF THE SEVEN PERMITTED COLORS.

That the quality of the lots of the seven permitted colors of Food Inspection Decision No. 76 offered on the markets of the world after the issuance of that decision was no better than that of the lots just reported, if as good, appears from the paper of E. G. Kohnstamm entitled, "Certified Food Colors: The Difficulties in the Way of their Manufacture," presented to the Seventh International Congress of Applied Chemistry held in London, May and June, 1909, and published in abstract form. This abstract reads as follows:

The coal-tar colors permitted under the food and drugs act of the United States are seven in number, which must be in a high state of purity. The author states that none of the colors on the markets of the world, at the time of testing, would meet these requirements.

Of these seven colors, 189 samples, from every possible source, and representing all the leading manufacturers, are here reported on, and the results of their examination reasonably establish the necessity of food color certification. Of these 189 samples, the worst were offered for food coloring purposes and seemed to be so offered because unfit for any other purpose.

Naphthol Yellow S.—Sixty-four samples examined, ranging in shade from clear bright yellow to a dirty brownish or green color. All contained Martius Yellow, some more than 1 per cent thereof; some contained as high as 2 per cent and even 3 per cent unconverted initial material or decomposition products. Forty-one contained excessive arsenic and 29 excessive heavy metals.

Orange I.—Twenty-eight samples examined; all contained decomposition products varying from a slight amount to over 50 per cent; free  $\alpha$ -naphthol was found in most samples; in 12 it was as high as 2 per cent; insoluble matters were as high as 1 per cent; lead to the extent of 0.5 per cent was found in one sample; shading by added colors and excessive amounts of arsenic; lead and iron were frequent.

Amaranth.—Thirty-eight samples examined; none were pure, and all contained arsenic in excessive amounts, and in one case as high as 0.1 per cent; all contained added color, principally an acid-violet. Iron as high as 0.1 per cent; insoluble matter as high as 1½ per cent; was most heavily loaded with salt of all seven colors.

Ponceau 3R.—Thirty-six samples examined; the purest of all colors tested; not toned; heavily loaded with salt; much insoluble matter was present; decomposition products were absent; iron, 0.01 per cent; 12 contained excessive amounts of arsenic and 14 contained excessive amounts of heavy metals.

Erythrosin.—Twelve samples examined; 10 were not erythrosin at all; of the other two, one was low in iodin and one contained arsenic.

Light Green S F Yellowish.—Thirteen samples examined; only one free from arsenic; nine contained lead or copper; one contained manganese; none were loaded.

Indigo Disulphonic Acid.—Eight samples examined; none were pure; the iron content was as high as 1.5 per cent; all were loaded with salt or Glauber's salt. One contained excessive amount of arsenic and two excessive amounts of heavy metals.

The difficulties consist in keeping the undesirable materials out of the dyes or in separating them from the crude dyes, or both.

In this connection the following statement made by Dr. E. Ludwig, of Vienna, at the International Congress of Medicine held in Budapest, August, 1909, may be of interest:

The author, at an order of a court, at the beginning of the seventies in the last century, examined approximately 200 samples of food products confiscated as suspicious and taken from numerous sales places of a then suburb of Vienna; these samples included solid confectionery, fruit sirups, spirits, etc. He found that more than 90 per cent of these things were colored with magenta and contained arsenic. In some of the sales places the preparation used for coloring, the so-called "couleur," was found, which proved to be a solution of magenta and in which there were contained 8 per cent of arsenic in the form of arsenous acid and of arsenic acid. According to the statements of a qualified dyestuff maker, this "couleur" was a mother liquor from magenta manufacture, which was very difficultly saleable and which, however, a conscienceless agent had talked onto ignorant producers of and dealers in foods.

Schacherl (p. 1046) says: "It should be required of all permitted coloring matters that they shall not contain substances which are harmful to health, or even suspicious, either in chemical union or as contaminations." The following pages (Section XV) show how closely this requirement has been met as a result of quality control on the part of the Department of Agriculture.

## XV. GUIDES IN DETERMINING DEGREE OF PURITY AND CLEANLINESS.

In view of the absence of any statements in the literature defining the purity of the colors physiologically examined with such accuracy that another could obtain with reasonable certainty the same degree of cleanliness, it became necessary to devise some guide, no matter how empirical, in the setting up of standards, tentative or otherwise.

As a first consideration it was held that, in view of the fact that all of the physiological work had been done with specimens of coal-tar colors of commercial purity, it would be reasonable to suppose that coal-tar colors produced in the purest form possible with present-day methods would certainly be as clean and as free from admixture as any of the commercial products subjected to physiological test. It was considered unreasonable to expect that increasing the degree of purity of these substances could in any way increase any harmful property possessed by them. Certainly in the case of Naphthol Yellows, where Martius Yellow is a usual contaminant, it can hardly be maintained that decreasing the amount of Martius Yellow would increase any harmful property which might reside in Naphthol Yellow S proper; in the case of Ponceau 3 R it could hardly be argued that any undisazotized base or decomposition products of diazo compounds tended to correct or counteract any harmful property that might reside in Ponceau 3 R proper; nor could it be maintained that Orange I free from uncombined alpha-naphthol was more harmful than Orange I, contaminated with alpha-naphthol, and so on through the list of the seven permitted colors. This point would not be raised had it not been pressed repeatedly by different persons as a serious objection to quality control and purity standards of the seven permitted colors of Food Inspection Decision No. 76.

With this rule in mind, and referring to the tabulated results of the analyses of the 30 specimens of the seven permitted colors just given, the items in the analytical statements will each be separately discussed.

Common salt and ether extractive.—Common salt is a legitimate component of commercial brands of coal-tar colors in so far as these coal-tar colors are obtained by the so-called "salting out" process. The coal-tar colors are recovered from solution by the addition of common salt, which has the peculiar property of separating the coal-tar color from the water solution as an undissolved solid. The coal-tar color so obtained will contain more or less salt, which, from a commercial manufacturing point of view, is therefore an unavoidable constituent. The amount of salt so accompanying the coal-tar color depends upon the amount added to the color solution, and this is greatest when complete exhaustion of the coal-tar color solution is attempted. It is a matter of common experience that, as a rule,

the coal-tar color first separating in the salting-out process is cleaner and less contaminated than the portions last separating. In an endeavor to recover as far as possible all the dissolved coal-tar coloring matter the manufacturer adds a large excess of salt, and this carries with it a large amount of organic matter not coloring matter, as can be seen from the preceding analyses, where a high salt content is almost always accompanied by a high ether extract content. The ether extract content is a measure of the dirt or organic impurities of the coloring matter.

The objection to an excessive amount of salt in coal-tar coloring matters, when they are to be used for food-coloring purposes, does not reside in the salt per se, but is due to the fact that a high salt content seems generally to be accompanied by an unnecessarily large amount of organic material not coloring matter, and nothing good is known of such material, which is more than likely to be harmful.

The acetate ether extract is supposed to represent that part of the organic material not coloring matter which has no, or only slightly, basic properties; the acid ether extract is supposed to represent that part of the organic material not coloring matter which has acid properties. In the case of Naphthol Yellow S the acid ether extract will contain all of the Martius Yellow, and very likely will consist substantially of it. In the case of Erythrosin, of course, no acid ether extraction was attempted because the color acid is itself ethersoluble.

The following tabulation shows the percentages of salt based on the amount of coloring matter present contained in 26 of the specimens before reported on, arranged under each of the seven permitted colors:

Naphthol Yellow S: 0.62; 1.04; 2.97; 2.99; 3.51; 3.64; 4.38; 7.38; 11.82; 31.53. (Average, 6.99.)

Ponceau 3 R: 27.16.

Orange I: 3.91; 11.19. (Average, 7.55.)

Amaranth: 16.20; 37.39; 39.97; 42.85; 64.37; 68.89. (Average, 44.95.) Light Green SF Yellowish: 0.04; 0.10; 1.64; 1.88. (Average, 0.92.) Indigo Disulpho Acid: 8.31; 8.35; 44.02. (Average, 20.23.)

The following figures give the same data for the acetate, acid, and total, ether extract:

Ether extractives.

#### ACETATE ETHER EXTRACT.

Naphthol Yellow S: 0.08; 0.10; 0.13; 0.14; 0.16; 0.16; 0.16; 0.19; 0.27; 0.37. (Average, 0.18.)

Ponceau 3 R: 0.38.

Orange I: 0.02; 0.69. (Average, 0.36.)

Amaranth: 0.03; 0.14; 0.19; 0.31; 0.32; 0.46. (Average, 0.24.)

97291°-Bull. 147-12-13

Light Green SF Yellowish: 0.02; 0.02; 0.05; 0.05. (Average, 0.035.)

Erythrosine: 0.11.

Indigo Disulpho Acid: 0.02; 0.02; 0.03. (Average, 0.03.)

#### ACID ETHER EXTRACT.

Naphthol Yellow S: 0.33; 0.44; 0.72; 0.77; 0.78; 0.94; 0.95; 0.97; 1.07; 1.16. (Average, 0.81.)

Ponceau 3 R: 1.13.

Orange I: 0.23; 0.25. (Average, 0.24.)

Amaranth: 0.05; 0.05; 0.12; 0.15; 0.19; 0.27. (Average, 0.14.) Light Green SF Yellowish: 0.05; 0.05; 0.05; 0.05. (Average, 0.05.)

Indigo Disulpho Acid: 0.04; 0.06; 0.15. (Average, 0.08.)

#### TOTAL ETHER EXTRACT.

Naphthol Yellow S: 0.43; 0.63; 0.86; 0.93; 0.94; 1.02; 1.10; 1.11; 1.43; 144. (Average, 0.99.)

Ponceau 3R: 1.51.

Orange I: 0.27; 0.92. (Average, 0.59.)

Amaranth: 0.08; 0.26; 0.36; 0.38; 0.59; 0.61. (Average, 0.38.)

Light Green SF Yellowish: 0.07; 0.07; 0.10; 0.10. (Average, 0.09.)

Erythrosin: 0.11.

Indigo Disulpho Acid: 0.06; 0.08; 0.18. (Average, 0.11.)

Insoluble matter.—The amount of insoluble matter (total, volatile, and nonvolatile) is a measure of the cleanliness of the materials used, as well as of the cleanliness of treatment during the manufacture of the coloring matter. The following tabulation shows the variations in these three figures for each of the 26 specimens of the 7 permitted colors, examined, as in the preceding cases:

### Insoluble matter.

#### VOLATILE INSOLUBLE.

Naphthol Yellow S: 0.00; 0.00; 0.00; 0.02; 0.02; 0.05; 0.13; 0.86; 0.97; 1.46. (Average, 0.35.)

Orange I: 0.39; 1.66. (Average, 1.03.)

Amaranth: 0.32; 0.51; 0.83; 1.10; 1.71; 1.93. (Average, 1.07.)

Light Green SF Yellowish: 0.16; 0.38; 0.22; 0.41. (Average, 0.29.)

Erythrosin: 0.50.

Indigo Disulpho Acid: 0.77; 1.23; 1.25. (Average, 1.08.)

## NONVOLATILE INSOLUBLE.

Naphthol Yellow S: 0.00; 0.00; 0.00; 0.00; 0.00; 0.00; 0.00; 0.03; 0.05; 0.10; 0.27. (Average, 0.05.)

Orange I: 0.23; 0.25. (Average, 0.24.)

Amaranth: 0.21; 0.54; 0.55; 1.39; 1.78; 1.98; (Average, 1.08.)

Light Green SF Yellowish: 0.00; 0.06; 0.64; 0.65. (Average, 0.34.)

Erythrosin: 0.22.

Indigo Disulpho Acid: 0.00; 0.59; 0.59. (Average, 0.36.)

#### TOTAL INSOLUBLE.

Naphthol Yellow S: 0.00; 0.00; 0.00; 0.05; 0.05; 0.12; 0.40; 0.86; 0.97; 1.51. (Average, 0.40.)

Orange I: 0.62; 1.91. (Average, 1.27.)

Amaranth: 0.53; 1.37; 1.65; 1.90; 3.69; 3.71. (Average, 2.14.) Light Green SF Yellowish: 0.22; 0.47; 0.81; 1.02. (Average, 0.63.)

Erythrosin: 0.72.

Indigo Disulpho Acid: 1.23; 1.36; 1.84. (Average, 1.48.)

Work subsequent to these analyses showed that the insoluble matter, particularly the nonvolatile insoluble matter, was greater in certain batches than in others, and that these amounts were in excess of the maximum amounts reported therefor. Examination showed that these increased amounts of insoluble matter were probably due to variations in the water delivered to the factories by their respective municipal water supplies, since it was noted that in two or more cases the batches containing an exceptionally large amount of nonvolatile insoluble matter were manufactured at a time when the city water supply was abnormally hard; this item, therefore, has been made in some instances a little more elastic than the preceding analyses would seem to justify.

Arsenic.—Examination of the preceding tabulations shows that 14 out of 30 specimens tested passed the Gutzeit test for arsenic, and

16 failed to pass.

Heavy metals.—Inspection of the preceding tables shows that 13 out of 30 specimens passed this test, and that 17 failed to pass.

Combined arsenic and heavy metals test.—Inspection of the preceding tables shows that 11 out of 30 passed both tests jointly; that 3 passed the arsenic test and failed to pass the heavy metals test; and that 2 passed the heavy metals test and failed to pass the arsenic test.

Moisture.—The amount of moisture, as inspection of the preceding analyses shows, is variable, and is a factor not under easy control. As long as the coloring matters submitted for foundation certification were in powder form and the analyses disclosed the actual percentage of coloring matter it was considered that any control of this item would involve an amount of labor and provide opportunities for friction wholly out of proportion to any benefit that at present could be realized therefrom. Therefore such control was not instituted, although it by no means follows that the time may not come when control of this item will be necessary.

Sulphur and sulphated ash.—These two determinations in the case of all the permitted colors, except Erythrosin, give a rough measure of the extent of the sulphonation and of the saturation of the sulphonic acids with sodium. It is not the function of these two items to exclude the isomeric modifications, nor was any test applied to such of these 30 specimens as were examined in this respect to determine the presence or absence of such products. The attempts made in this direction are of a later date than the analyses already reported, and are indicated on page 210 of this report.

With these data available, and with the general rule stated at the opening of this section in mind, it was regarded as the proper course, when determining upon standards, to require that each color should pass the combined heavy metals and arsenic test of the United States Pharmacopæia, and in other respects should be as clean and as high-grade as the best of each class given in the preceding analyses. Just how closely it was possible to adhere to this rule will be shown later.

With respect to the arsenic requirement, there has been a great deal of discussion brought on by those interested in the manufacture of these colors. It was protested that colors could not be made uniformly and continuously under manufacturing conditions, which would contain an amount of arsenic so small that a quantity of the product containing 2 grams of the coloring matter in question would not respond to the Gutzeit test in the United States Pharmacopæia. The results above given with respect to conformity or nonconformity to the Gutzeit test, were, with a great deal of justification, not regarded as conclusive, because, as has been previously stated, there was no certainty that the amount of coloring matter actually taken did represent 2 grams. In view of the fact that the United States Pharmacopæia prescribes for the only coal-tar color in it, namely, Methylene Blue, that it shall be so free from arsenic that 2 grams fail to respond to the Gutzeit test, it was considered perfectly justifiable to adhere to that requirement until it could be conclusively shown that it was unreasonable and incapable of attainment. Results described in the pages that follow show that it has been possible to make all the 7 permitted colors of Food Inspection Decision No. 76 so free from arsenic that they comply with the Pharmacopæial test.

With respect to the heavy metals, no deviation was necessary for the nonferrous metals from the test of the Pharmacopæia; for iron, however, it was found necessary to increase the limit to substantially 0.005 part per 100 of actual coloring matter. The reason for this is that at one time or another all of the seven permitted coloring matters in the course of their manufacture come in contact with, or are contained in, vessels of iron, and it seems to be almost impossible to keep iron out to an extent which would bring the color within the pharmacopæial test. It has been shown that there were 13 colors on the market in 1907 which contained so little iron that they failed to respond to the heavy metals test of the Pharmacopæia in that respect; but here again the same criticism holds good as in the arsenic test, that there is no certainty that the amounts taken for the heavy metals tests were equivalent in each case to 1 gram of actual coloring matter, and subsequent experience seems to confirm the correctness

of that criticism; therefore the rule for cleanliness of product, as just given, has been modified in that respect and to that extent.

That it was only fair and reasonable to expect considerable improvement in the cleanliness and purity of food colors was made evident by an examination of two substances sold in the United States in large quantities for the purpose of making a very cheap red coal-tar coloring matter, which is used in many of the cheapest coloring operations, for paints, inks, etc. These substances are paranitranilin and betanaphthol. They were found in the United States market in such a condition of cleanliness and purity that had they been suitable for use in foods no objection could be raised against them on these scores. They complied with the requirements of the United States Pharmacopæia with respect to freedom from arsenic and all heavy metals, inclusive of iron. The significance of this lies in the fact that all the raw materials entering into the manufacture of paranitranilin and betanaphthol also enter into the manufacture of the seven permitted colors of Food Inspection Decision No. 76, and that the only source of arsenic in food colors, and probably the only way in which iron could be introduced into them, is by way of the materials entering into the manufacture of paranitranilin and betanaphthol, and since it has been shown to be commercially possible to keep those bodies out of paranitranilin and betanaphthol, and since there is no occasion whatever for arsenic or iron or other heavy metal being present in any of the materials used in the manufacture of the seven permitted colors of Food Inspection Decision No. 76, over and above the materials used in the manufacture of paranitranilin and betanaphthol, there was every reason for believing that the seven permitted colors of Food Inspection Decision No. 76 could ultimately be manufactured and marketed in the same degree of cleanliness and purity that paranitranilin and betanaphthol are marketed; in other words, that food colors could be made as clean and as pure as paint colors, a condition not existing in the food-color market of the United States in the summer of 1907.

The results of the control exercised by the Department of Agriculture over the quality of food colors, as compiled in the following section, fully justify such expectations, as well as the aim to make coal-tar colors when used for food purposes of the same high degree of cleanliness and purity as when they are to be used as drugs or as they actually are when used for the manufacture of paints and printer's inks; that is to say, that the coal-tar color used in a colored food should be as clean as the coal-tar color used in making the ink on the label of such colored food, the very reverse of the situation existing prior to the quality control now established.

# XVI. ANALYSES OF CERTIFIED LOTS OF PERMITTED COLORS, 1909-10.

### TABULATION OF RESULTS.

The analytical results obtained on 74 batches of certified colors, totaling upward of 32,000 pounds, are given in the following tables. The analytical results are the work of the New York Food and Drug Inspection Laboratory in the course of checking up the analytical results certified to in foundation and in supplementary certificates. The period covered by these examinations is approximately from July, 1909, to January 1, 1910.

## Naphthol Yellow S.

[Figures calculated to 100 parts pure color and arranged in the order of their size. Figures on the same horizontal lines do not refer to the same samples; the dash line shows the location of the average of each column.]

	Insolu	ıbles.	]	Ether ex	tractives	3.	Com-	Cont	So-	Cal
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	Sul- phur.	dium.	Cal- cium.
12	0.170 .130	0.100 .078	0.036 .026	0.016 .007	0.067 .065	0.088 .073	4.06 2.26	9.23 8.99	13.27 13.21	0.13 .13
3 4	.097	.050	.024	.006	.042	.072 .064	1.68 1.41	8.98 8.96	12.92 12.90	.05
5	.082	.040	.019	.005	.036	.061	1.30	8,89	12.89	
6	.079	.036	.018	.005	. 034	.057	.63	8.86	12.89	
7 8	.070	.032	.016 .012	.004	.031 .026	.056	.63 .48	8.85 8.83	12.87 12.87	
9	.055 .050 .050 .030 .030	.010	.011 .010 .007 .006	.000 .000 .000 .000	.025 .025 .020 .018	.048 .045 .045 .045 .044	.47 .25 .24 .19	8.71 8.70 8.62		
Average	.076	.043 4 6	.017 6 6	.004	.035 5 7	.051 8 5	1.06 5 8	8.87 5 6	12.93 2 8	.09

#### Ponceau 3R.

[Figures calculated to 100 parts pure color and arranged in the order of their size. Figures in the same horizontal line do not refer to the same sample; the dash line shows the location of the average of each column.]

27	Insol	uble.	Е	ther ex	tractive	es.	Com-	So-	G1	\ a_	G-1	Boiling
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	dium sul- phate.	Sul- phur.	So- dium.	Cal- cium.	point of cumi-din.
12	0.55 .42	0.29	0.166 .163	0.034	0.019	0.216	5.72 5.67	0.06	13.03 12.99	9.51 9.49	0.18	°C. 222-235 222-235
3	.41	.17	. 161	.032	.018	.201	5.57	.00	12.95	9.31	.14	222-235
4	.27	.12	.072	.009	.018	.098	5.00	.00	12.88	9.31	.14	220-230
5	.17	.11	.053	.006	.017	.073	4.12	.00	12.80	9.23	.12	220-230
6	.17 .16 .14 .10	.11 .09 .07	.052 .038 .038	.002 .002 .000	.017 .016 .014	.072 .058 .057	3.73 3.71 3.03 2.53	.00 .00 .00	12.80 12.77 12.64	9.16 9.12 9.07 8.74	.12	220–230 220–229 225–228 216–226
Average Above Below	.266 4 5	.15 3 5	.093 3 5	.015	.017	.124 3 5	4.34 4 5	.01	12.83 4 4	9.21 5 4	.14	221-231 3 6

## Orange I.

[Figures calculated to 100 parts pure color, and arranged in the order of their size. Figures in the same horizontal line do not refer to the same sample; the dash line shows the location of the average of each column.]

	Insol	uble.	1	Ether ex	tractives		Com-	So-	Co. 1	9-	0-1
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	dium sul- phate.	Sul- phur.	So- dium.	Cal- cium.
1	0.39	0.18	0.224	0.093	0.08	0.360	3.13	0.22	9.26	6.82	0.29
2 3	.38	.08	.220	.060	.077 .067	.309	3.13 3.04	.10	9.23 9.12	6.76 6.63	.24 .24
4	.23	.05	.207	.035	.055	.300	2.36		9.10	6.62	.22
5	.23	.04	.181	.035	.048	.269	1.98		9.06	6.54	.05
6	.16	.04	.177	.020	.039	.267	1.91		9.06	6.48	.04
7	.15	.03	.174	.018	.036	.246	1.90		9.05	6.32	
8	.11	.03	.124	.015	.029	.224	1.41		9.02	6.21	
9	.10	.02	.107 .097	.011	.016	.175 .124	1.05 1.01		8.96 8.15		
Average Above Below	.21	.06 3 6	.172 7 3	.034 5 5	.050 4 5	.258 6 4	2.09 4 6	.03 1 1	9.00 8 2	6.55 4 4	.18 4 2

#### Amaranth.

<sup>[</sup>Figures calculated to 100 parts pure color and arranged in the order of their size. Figures on the same horizontal lines do not refer to the same samples; the dash line shows the location of the average of each column.]

	Insol	uble.	)	Ether ex	tractives		Com-	Sul-	Sodi-	Calci-
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	phur.	um.	um.
1 2 3	0. 43 . 39 . 29	0.33 .30 .19	0.090 .088 .069	0.030 .017 .009	0.059 .017 .015	0.158 .101 .100	4.18 4.16 3.83	16. 04 15. 80 15. 63	14. 22 14. 19 12. 10	0.20 .19 .17
4	. 23	.15	. 047	.007	.014	.060	3. 73	15.61	11. 47	.14
5	. 21	.10	.042	.007	.012	.056	3.67	15.59	11.43	.14
6	. 13	.10	.035	.005	.012	. 053	3.08	15.58	11.43	.13
7	.12	.06	.034	.005	.012	.052	3.02	15.57	11.42	. 13
8 9	.12	.05 .05	.034	.005	.010	.051 .048	2. 94 2. 70	15.57 15.56	11.38 11.33	.12
10. 11. 12. 13.	.11 .11 .11 .11	.04 .04 .040 .036	.031 .031 .031 .027	.004 .004 .004 .004	.008 .008 .007 .006	.042 .042 .040 .039	2. 62 2. 21 2. 12 2. 12	15.54 15.52 15.52 15.52	11.32 11.31 11.29 11.27	.12 .11 .09 .09
14. 15. 16. 17. 18. 19. 20. 21.	.092 .09 .09 .089 .08 .070 .07	.03 .03 .025 .02 .02 .02 .02	.026 .026 .025 .021 .019 .018 .012	.004 .003 .002 .001 .000 .000	.006 .005 .005 .004 .003 .003	.038 .038 .038 .035 .035 .029	2. 04 1. 94 1. 94 1. 91 1. 91 1. 23 1. 22	15. 47 15. 43 15. 41 15. 41 15. 41 15. 40 15. 12 15. 12	11. 27 11. 23 11. 21 11. 18 11. 15 11. 14 11. 10 11. 01	.09 .09 .08 .07 .00
Average	.148 5 16	.083 6 14	. 037 5 15	.006 5 15	.011 7 12	.056 4 15	2.63 9 11	15.51 13 8	11. 59 3 18	.012 7 11

## Light Green SF Yellowish.

[Figures calculated to 100 parts pure color and arranged in the order of their size. Figures in the same horizontal line do not refer to the same sample; the dash line shows the location of the average of each column.]

Nhan of	Insol	uble.	]	Ether ex	tractives		Com-	Sodi-	Sul-	n-4:	C-1-i
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	um sul- phate.	phur.	Sodi- um.	Calci- um.
1	0.07	0.034	0.058	0.011	0.011	0.076	4. 15	0.00	12.69	8.03	4. 52
2	. 045	.02	.041	.007	.009	. 054	. 64	.00	12.27	7.99	.10
3 4 5	.03 .02 .02	.01 .01 .01	.034	.006	.008	.053	.515 .43 .37	.00	12.07 11.88 11.88	7. 99 7. 94 7. 73	.09 .06 .05
6	.01	.01						.00		1.56	
Average Above Below	. 033	.017	.041	.006	.008	.056	1. 221 1 4	.00	12.16 2 3	6.87 5 1	.96

### Erythrosin.

[Figures calculated to 100 parts pure color and arranged in the order of their size. Figures in the same horizontal line do not refer to the same sample; the dash line shows the location of the average of each column.]

	Insol	uble.	:	Ether ex	tractives		Com-	Sodi-	Sodi-	
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	um sul- phate.	um.	Iodin.
1	0.13 .09	0.09 .04	0.039	0.017 .009		0.051 .048	1.13 1.00	0.00	5. 42 5. 35	57. 45 56. 46
3	. 059	.024	.037	.008		.047	. 48	.00	5. 33	56. 40
4	.05 .04 .04	.01 .01 .01	.034 .033 .033	.002 .002 .000		. 037 . 035 . 035	. 23 . 09 . 09	.00	5. 29 5. 15 5. 11	56. 07 55. 98 55. 97
7	.04	.01	.027			.027	.09 .09 .036	.00	3.85	55. 94 55. 90 55. 88
Average. Above. Below.		.028	.035	.006		.040	.360 3 6	.00	5. 07 6 1	56. 23 3 6

## Indigo disulpho acid.

[Figures calculated to 100 parts pure color and arranged in the order of their size. Figures on the same horizontal lines do not refer to the same samples; the dash line shows the location of the average of each column.]

N	Insol	uble.		Ether ex	tractives		Com-	Sodi-	C1	a.a:	G-1-1
Number of batches.	Total.	Inor- ganic.	Neu- tral.	Alkali.	Acid.	Total.	mon salt.	um- sul- phate.	Sul- phur.	Sodi- um.	Calei- um.
1	0.50	0.28	0.140	0.087	0.099	0.320	6. 77	16.12	13. 45	10.13	0.45
2	. 46	. 26	. 134	.061	.038	. 217	3.60	10. 22	13. 43	9.90	.13
3	. 45	.25	.102	.029	.026	.143	3.21	4.79	13. 42	9.64	. 13
4	. 43	. 24	.088	.015	.024	. 141	2. 89	.00	13. 39	9.60	.05
5 6	.42	.22	.075	.013	.016	.126	1.85 .40	.00	13. 22 13. 15	9. 13 8. 54	
Average Above Below	. 43	. 235	.108	.041	.041 1 4	.189	3.12	5. 19 2 4	13.34 4 2	9. 49 4 2	. 19

#### COMPARISON WITH ANALYSES MADE IN 1907.

A comparison, as far as possible, of these figures with the corresponding data previously given for samples on the market in the summer of 1907 is made in the following table. The figures in this new table show the value of the fraction obtained by dividing the old average figure by its corresponding new average figure—that is, they show how many times greater the old average figures are than the corresponding new average figures. Italics indicate those items in which there has been a retrogression in the new figures as compared with the old; all the other figures represent an advance. It will be noted that comparisons are not made for Ponceau 3R and Erythrosin. This comparison was omitted because there was but one sample of Ponceau 3R examined and a partial analysis of one sample of Erythrosin in the old work.

Ratio	of	average	figures of	1907	to	those	for	the	certit	fied	colors.	1909.
10000	V.1	uveruge	Jug will be of	1001	00	CIECUSC.	, 01	0100	corve	0000	000010,	1000.

			Insoluble.		Et	her extrac	ts.
Color.	Salt.	Total.	Volatile.	Nonvol- atile.	Nonacid.	Acid.	Total.
Naphthol Yellow S. Orange. Amaranth Green. Blue.	4.88 3.54 15.07 .66 8.05	4.44 9.07 11.26 31.00 3.29	9. 02 20. 60 21. 40 29. 00 5. 4	0.67 3.00 7.71 17.00 1.56	7.50 1.57 4.07 .65 .15	21.89 4.53 10.77 6.25 3.48	15.97 2.26 5.00 1.67 .68

## These retrogressions are:

- 1. Salt in the Green.
- 2. Nonvolatile insolubles in the Yellow.
- 3. Nonacid ether extract in the Green.
- 4. Nonacid ether extract in the Blue.
- 5. Total ether extract in the Blue.

With respect to the first of these retrogressions there is this to be said: One lot of Green had apparently been purified by precipitation with salt, since it was in every other respect of a high quality—that is, it was free from arsenic and heavy metals within the pharmacopœial test, and its ether extractives were very satisfactory. The other lots of Green examined had apparently not been made in this manner.

With respect to the second retrogression, the probable explanation is that some of the lots were made during a period when the municipal water supply was excessively hard, as before explained. The remaining three retrogressions are probably due to the fact that the old methods of analysis used were not so accurate as the methods later employed and hereinafter described (pp. 223, 225).

It will be noted that in three of the six batches of Indigo disulpho acid reported there is no sodium sulphate; whereas in the other three batches the amount of this substance is as high as 16.12. The reason for this is that in the early stages of the work the results obtained

by the manufacturers pointed in the direction of the impossibility of getting rid of all the sodium sulphate; but later and more extended work showed this conclusion to be an error. There is therefore no good reason for permitting sodium sulphate as a contaminant in this or any other of the seven products.

## CONFORMITY OF ANALYTICAL DATA WITH THEORETICAL COMPOSITION.

The seven tables following, likewise based upon the 74 Government analyses, show the conformity or nonconformity, as the case may be, of these samples with their theoretical composition. The second and third columns show the percentage based on theory of the contained sulphur and sodium in the case of all colors except Erythrosin, where the figures for iodin are substituted for those of sulphur. The fourth column shows the actual ratio of sodium to sulphur or iodin, as the case may be, and the fifth column gives the percentage of the figure of column 4 based on the theoretical value which is given in parentheses at the head of columns 4 and 5. The summary gives the maximum, minimum, and average of each column. Taken as a whole the figures are fairly satisfactory and show a reasonable conformity of the actual product to theoretical requirements, although there seems to be considerable room for improvement, which no doubt can be achieved in time.

Percentages based on theoretical composition.

NAPHTHOL YELLOW S.

Number of batches.	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (1.4379).$ Value. Per cent.		Number of batches.	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (1)$ Value.	Per cent.
1 2 3 4	Per cent. 103. 120 96. 312 97. 205 97. 542	Per cent. 103. 10 99. 068 100. 000 99. 534	1. 4377 1. 4791 1. 4792 1. 4707	99. 983 102. 860 102. 870 102. 270	10 11 12 13	Per cent. 98. 880 98. 658 100. 330 100. 100	Per cent. 101.06 101.22 100.15 100.00	1. 4576 1. 4631 1. 4353 1. 4363	101.36 101.74 99.826 99.890
6 7 8 9		100. 15 103. 50	1. 4498 1. 4692	100. 80 102. 18	Average Max Min	99. 101 103. 120 96. 312	102, 212 103, 500 99, 068	1. 4578 1. 4792 1. 4353	101.378 102.87 99.826

#### PONCEAU 3R. Na =(0.71905).=(0.71905).Number Number Sulphur. Sodium. Sulphur. Sodium. of batches. batches. Value. Per cent. Value. Per cent. Per cent. 101. 920 101. 700 Per cent. Per cent. Per cent. 0.74296 103.33 0.69138 96.17398.693 99.308 97.454 93.617 98. 927 97. 749 98. 174 . 73679 102.48 97.351 100.460 .69992 99.144.70518 98.080 99.256 98.738 .71648 100.140 Average. 103.33 96.173 97.095 .70936 98.662 99.918 Max.... 100.46 101.920 .74296 97.454 93.617 6.... 99.841 99.783 .71892 Min .... 101, 16 98, 693 99, 783 .72730

## Percentages based on theoretical composition—Continued.

## ORANGE I.

Number	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (0$	<b>).</b> 71905).	Number	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (0$	.71905).
batches.			Value.	Per cent.	batches.			Value.	Per cent.
1 2 3	88. 972 100. 76 101. 09 99. 135	100. 44 103. 72 102. 53	0.81226 .73889 .73002	112.97 102.77 101.53	8 9 10	98. 471 99. 563 99. 342	85. 901 99. 241 98. 330	0.70066 .71710 .71208	97. 454 99. 740 99. 042
5 6 7	99. 135 97. 816 98. 799	94. 234 100. 610	. 69309 . 73243	96. 391 101. 90	Average . Max Min	98.308 101.09 88.972	99.376 103.72 95.901	.72957 .81226 .69309	101.475 112.97 97.454

## AMARANTH.

1	96.795	105.76	0.78519	109.21	14	97.800	98.863	0.72640	101.03
2	98.176	99.037	.72488	100.82	15	97.499	99. 911	. 73646	102.43
3	94.975	124. 29	. 94048	130.80	16	97.614	98. 950	.72843	101.32
4	94. 975	124.04	. 93849	130.53	17	97.499	99.476	. 73325	101.98
5	100.750	96.240	. 68637	95.470	18	97.863	98. 689	.72465	100.78
6	99. 246	99. 911	.72341	100.62	19	98.052	98. 393	. 72106	100.05
7	97.174	100.260	.72454	100.78	20	97.800	98. 514	.72383	100.68
8	96.929	99.827	.74011	102.94	21	97.735	97.725	.71852	99.914
9	97.926	97.465	.71520	99.476	13				
10	96.975	97. 990	.72744	101.18	Average.	97.467	101.347	.74707	103.90
11	96.733	97.389	. 72338	100.62	Max	100.750	124. 29	.94048	130.80
12	97.487	98. 514	. 72615	100.99	Min	94.975	96. 240	.68637	95.470
13	96.798	97.028	. 72030	100.18					
		U .							

### LIGHT GREEN SF YELLOWISH.

1	102.85				6	104. 50	95. 547	0.65783	91. 495
3 4 5	106. 23	93.018 96.629	0.60911 .65358	84. 722 90. 911	Average. Max Min	109.87	96.629	67256	90. 167 93. 540 84. 722

#### ERYTHROSIN.

Number of	Iodin. Sodiun		$\frac{\text{Na}}{\text{I}_2} = (0.090855).$		Number	Iodin.	Sodium.	$\frac{\text{Na}}{\text{T}_2}$ =(0.090855).	
batches.			Value.	Per cent.	batches.			Value.	Per cent.
1 2345 67	Per cent. 96.915 97.051 99.593 97.780 97.209 96.983 97.035	Per cent. 73.473 100.94 98.277 97.521	0.068873 .094497 .089639 .090604	75. 805 104. 00 98. 663 99. 724	8 9 Average. Max Min	Per cent. 96.879 97.886 97.952 99.593 96.879	Per cent. 102.09 103.43 96.777 103.43 73.473	.095741 .095995 .090204 .095995 .068873	105. 37 105. 65 99. 146 105. 65 75. 805

## Percentages based on theoretical composition—Continued. INDIGO DISULPHO ACID.

Number of batches.	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (0$	.71905).	Number of	Sulphur.	Sodium.	$\frac{\text{Na}}{\text{S}} = (0$	.71905).
			Value.	Per cent.	batches.			Value.	Per cent.
1	Per cent. 97.670 95.636	Per cent. 103. 330 98. 279	0.71482 .69430	99.423 95.669	6	Per cent. 97.818	Per cent. 109. 040	. 75315	104.750
3 4 5	97.600 96.148 97.382	103.760 91.926 106.560	.71832 .64598 .73934	99. 910 89. 848 102. 830	Average. Max Min	97.042 97.818 95.636	102.149 109.040 91.926	.71099 .75315 .64598	98.738 104.750 89.848

### ARSENIC DETERMINATIONS ON 86 BATCHES.

During the course of this work it became necessary to determine the exact amount of arsenic contained in 86 of the various batches of certified colors; for this purpose the arsenic method of Seeker and Smith (see p. 212) was devised. These results are expressed in the number of parts of color containing one part of metallic arsenic (As). The numbers in parentheses indicate the number of specimens of the quality indicated.

Naphthol Yellow S (17 specimens).—770,000, 833,000, 1,250,000 (2), 1,428,000 (2), 1,666,000 (3), 2,000,000 (6), 3,333,000 (2).

*Ponceau* (15 specimens).—588,000, 625,000, 667,000, 714,000 (2), 769,000 (2), 1,000,000 (2), 1,111,000, 1,250,000, 1,666,000 (2), 2,000,000, 2,500,000.

Orange (7 specimens).—200,000, 250,000, 588,000, 1,000,000, 1,111,000, 1,429,000, 5.000.000.

Amaranth (27 specimens).—909,000, 1,000,000, 1,111,000, 1,250,000 (2), 1,438,000 (3), 2,000,000 (4), 2,500,000 (4), 3,333,000 (9), 5,000,000 (2).

Green (8 specimens).—166,000, 200,000, 370,000 (2), 500,000, 666,000, 770,000, 833,000.

Erythrosin (7 specimens).—2,000,000 (2), 5,000,000 (2), 10,000,000 (2), 20,000,000 (1).

Blue (5 specimens).—285,000, 666,000, 1,428,000, 3,333,000 (2).

The United States Pharmacopæia test for arsenic is sensitive to 0.005 mg of arsenious oxid (As<sub>2</sub>O<sub>3</sub>) which on a sample containing 2 grams of actual or real color would amount to one part of arsenious oxid in 400,000 of color; calculated to metallic arsenic, the basis employed in the foregoing, this would mean 528,000 parts of color for each one part of metallic arsenic.

There are, therefore, in the foregoing 86 lots of certified colors 8 which did not comply with that requirement, namely:

Orange (2 specimens).—200,000 and 250,000.

Green (5 specimens).—166,000; 200,000; 370,000 (2); 500,000. Blue (1 specimen).—285,000.

The reason for this discrepancy is that in the preparation of the samples for analysis by the United States Pharmacopæia test a loss of arsenic ensued, which is avoided in the Seeker-Smith method now employed in making these determinations. Had the existence

of this discrepancy been proven at the time the first analysis was made certification would have been denied to the eight lots above mentioned.

These results further show the position taken early in the work by several of the manufacturers—that a requirement of not more than 1 part of metallic arsenic in not less than 264,000 parts of coal-tar color could not be complied with on a commercial scale—to be untenable; on this basis only 4 out of the 86 lots examined would have been excluded. Further, the position of some manufacturers and dealers that the arsenic requirement ought, for practical manufacturing reasons, not to be more rigorous than one part of metallic arsenic in 26,400 parts of color, or 1 part of arsenious oxid (As<sub>2</sub>O<sub>3</sub>) per 20,000 parts of color, is not borne out by the data.

## SUGGESTED REQUIREMENTS FOR CERTIFIED COLORS.

Although the material embodied in this report gives a very good idea of the composition and quality of substantially 30 different lots of permitted colors prior to the issuance of Food Inspection Decisions Nos. 76 and 77, and of 74 lots of certified colors, yet these data are hardly sufficient to furnish a basis for standards with which each color specimen must comply in detail. The fitness or unsuitability of any lot has been determined by the examination of the analytical data obtained thereon in the Food and Drug Inspection Laboratory at New York; such examination has been applied to the particular color under investigation with respect to its general relationship to the results theretofore achieved. If in some minor quality, as, for example, freedom from salt, the sample was not up to what had been previously accomplished, but in a major quality, as for example, ether extractive, it was equal to or better than what had been previously accomplished, and the pharmacopeial tests for arsenic and heavy metals were satisfied with the exception of iron. and the amount of iron was within the limit previously stated, 0.005 per cent, and the other factors showed a fairly close conformity. such a defect as its salt content would not act as a bar to the passing of the lot; however, no matter how good a color might be in respect to such determinations as ether extractive, if it failed to comply with the United States Pharmacopæia requirement for arsenic or for heavy metals it was not accepted.

These arsenic results have been tabulated to show the distribution of arsenic content (As<sub>2</sub>O<sub>3</sub>); the numbers at the top are the Green Table numbers; the numbers in the body of the table indicate the number of specimens of the arsenic content stated in the first column; the last column shows the totals of all colors of the arsenic content (As<sub>2</sub>O<sub>2</sub>) corresponding thereto.

Arsenic content of 86 lots of certified colors.

Arsenic		Green Table numbers.								
content.	4	56	85	107	435	517	692	Total.		
1 part in—  i 126, 200  1 151, 000  1 151, 500  1 159, 300  1 215, 900  1 228, 300  1 228, 300  445, 400  443, 500  504, 600  505, 300  540, 900  688, 700  757, 500  688, 700  7757, 500  1, 081, 000  1, 082, 000  1, 082, 000  1, 1, 083, 000  1, 1, 083, 000  1, 1, 083, 000  1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1	1 1 1 2 2 2	1 1 1 2 2 1 1	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	1 1 1 1 2 3 3 4 4 9 2	11 11 12 11	2 2 2 1	1	1 1 1 1 2 1 2 2 2 2 2 2 2 2 1 4 3 5 3 1 3 5 1 3 5 1 1 3 5 1 1 3 5 1 1 3 5 1 3 1 3		
	17	15	7	27	8	7	5	86		

<sup>&</sup>lt;sup>1</sup> The certification of these lots was due to an unknown source of error in the analytical method; the analyses made at that time (July, 1909), showed less than 1 part of As<sub>2</sub>O<sub>3</sub> per 400,000; see also page 204.

It is therefore somewhat premature to attempt to define rigidly the requirements for composition and purity for colors until a sufficient number of analyses is available to permit a hard-and-fast line to be drawn for each item as required for each color. Until that time the decision as to whether or not a certain color shall be certified must rest with the Department of Agriculture. However, the following requirements are tentatively suggested as being commercially practicable. It should be clearly understood that the tentative requirements here stated are based on the results of actual control, and are not any more searching or numerous than are the requirements for many if not most of the coal-tar dyes or their component parts in the industrial arts, particularly for the various kinds of paint, varnish, and ink making. While it may be that some of the tentative requirements herein defined necessitate the expenditure of considerable work and time, yet that is also true of some of the commercial requirements. Since manufacturers of cheap paints, varnishes, inks, and the like, find it wise and necessary carefully to control the quality of the coal-tar dyes or their component parts which they use, it can not be less wise or necessary to extend the same kind of quality control to those coal-tar dyes intended and sold for human consumption as food. This stand is fully justified by the quality of the coal-tar dyes offered as food colors on the United States markets as described in the foregoing pages. That such control is not only practical, but practicable, is fully proved by the fact that more than 20.5 tons (41,000 pounds) of coal-tar food dyes have been so controlled, examined, and certified under the food inspection decisions hereinbefore mentioned.

The figures are expressed in parts per hundred of actual color contained in the dye and not in parts per hundred of the total substance. The numbers at the head of the columns are the numbers in the Green Tables.

Tentative limits of composition suggested for permitted colors.

(Parts per	hundred	of	actual	color.	)
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Determinations.	Green Tables numbers.								
Determinations.	4	56	85	107	435	517	692		
Insolubles: Total. Nonvolatile on ignition Ether extractives: Neutral. Alkaline Acid. Common salt.	0.070 .040 .017 .004 .050 .600	0.270 .150 .090 .015 .017 5.000	0.250 .050 .150 .035 .050 2.000	0.130 .050 .030 .005 .010 2.500	0.030 .010 .040 .006 .008 .600	0.060 .020 .035 .002	0. 450 . 250 . 100 . 030 . 150 3. 000		

In addition to these there are the following requirements applicable to all colors:

- 1. The absence of admixed dye must be convincingly demonstrated by suitable test.
- 2. Arsenic.—Test 17 of the United States Pharmacopæia, 1900, applied to so much of the specimen as represents 3.5 grams of actual dye must give a negative response. Such negative result must be reenforced by a check test identical with the test on the dye with the addition of 0.005 mg of arsenic (As<sub>2</sub>O<sub>3</sub>) to the dye prior to treatment, and this check test must produce a positive result for the presence of arsenic. Ignitions in the preparation of the material for the test must be made in porcelain.

Any other mode of testing which is demonstrated to be capable of detecting 0.005 mg added arsenic ( $As_2O_3$ ) in so much of the specimen as represents 3.5 grams of actual color will, of course, be accepted. This quantity, however, is only tolerated tentatively pending further investigations relative to the complete, or practically complete, elimination of arsenic from foods, especially those which are largely used by children, such as candies.

- 3. Heavy metals.—Test 121 of the United States Pharmacopæia as revised May 1, 1907, using so much of the specimen as represents one part of actual dye must give a negative response for all metals except iron, which may be present in amounts not in excess of 0.005 per cent of iron based on the dye actually present in the specimen.
- 4. None of the dyes offered for certification shall contain any Glauber's salt or sodium sulphate in any form, nor shall they contain any added sugar, dextrin, or other loader, filler, or reducer for any purpose whatsoever, and convincing proof of the absence of any or all of them must be submitted.

- 5. The ether extractives are to be made successively upon water solutions of the dye, first neutral, then made alkaline with caustic soda, and then made acid with hydrochloric acid, using washed or sodium, dried ether.
- 6. The sulphur content of the sulphur-containing dyes must agree substantially with the theoretical; likewise the sulphated ash figures of all must agree substantially with the theoretical; variations between these two sets of figures, as long as they are consistent with each other, will not be reason for rejection.
- 7. In the case of No. 4 proof must be submitted showing that the specimen is of the sodium or potassium variety, and if it is a mixture of these two varieties the proportion of each present in the mixture must be stated.
- 8. In the case of No. 56 the crude cumidin employed may have a boiling point of from 220° to 230° C., and may be liquid or solid; the absence of any compound of S or G salt must be convincingly shown.
- 9. In the case of No. 85 convincing proof must be submitted that beta-naphthol orange if present at all is present in an amount not in excess of 5 per cent of the coaltar dye present.
- 10. In the case of No. 107 the absence of any compound of S or G salt must be convincingly shown.
- 11. In the case of No. 435 the product should be free from calcium; convincing proof of absence of No. 434 must be submitted.
- 12. In the case of No. 517 the actual dye must contain not less than 56 per cent of iodin (sodium basis) and must not contain any other halogen; the kind and amount of metallic base, whether sodium, potassium, or the like, must be shown.
- 13. In the case of No. 692 the absence of indigo monosulphonic acid and of nonsulphonated indigo must be convincingly shown.
- 14. Each foundation certificate must be filed in duplicate, but need not be executed in duplicate and must contain a summarized or tabulated statement of all the quantitive results contained in the certificate, also a tabulation or summary of the qualitative tests made, together with the results of such tests, all stated on one sheet, so that the certificate will bear within itself its own summary and conclusions.
- 15. The fundamental analytical data must be given with such fullness as to permit efficient checking of the calculations, and the arithmetical operations performed should be indicated wherever needful to avoid confusion, or to facilitate the work of the checking chemist, or make the meaning of the certificate more plain.

There are freely quoted in the United States market two substances, paranitranilin and betanaphthol, which are subject to much competition, the prices for which, wholesale, are not far from 25 cents and 9 cents, respectively. A specimen of each has been examined, and, as before stated, they have both been found to be of such purity, with respect to arsenic, heavy metals, and general cleanliness, that had they been capable of use in food products, no objection against their use on this score could reasonably be raised; certainly no such objection could be successfully maintained.

The following table discloses the chemicals entering into the manufacture of paranitranilin and of betanaphthol, as well as of each of the seven permitted colors; the ingredients numbered 1 to 7 are used in the manufacture of these two substances, as well as in the seven permitted colors, as indicated by the "x" entries; ingredients 8 to 20 are used only in the manufacture of the seven permitted colors, and not in paranitranilin and betanaphthol.

Comparison of chemicals entering into the composition of the seven permitted colors and of paranitranilin and betanaphthol.

Determinations.	4. Nap- thol Yellow S.	56. Pon- ceau 3R.	85. Orange I.	107. Ama- ranth.	435. Light Green.	517. Ery- thro- sin.	692. Indigo disul- pho acid.	Parani- trani- line.	Beta- naph- thol.
1. Naphthalene 2. Benzol. 3. Sulphuric acid 4. Nitric acid 5. Metallic iron 6. Acetic acid 7. Caustic soda or potash	x x	x x x x x x	x x x x x x x	X X X X X	X X X X X	x x x	x x x	x x x x x	x x x
8. Carbonate of soda. 9. Common salt. 10. Wood or methyl alcohol. 11. Lime. 12. Hydrochloric acid. 13. Sodium nitrite. 14. Ethyl chlorid. 15. Peroxid of lead or of manganese. 16. Phosgene. 17. Iodin. 18. Metallic mercury. 19. Ferric chlorid. 20. Chlorin.	X	X				x	x x x x 10	5	

With respect to the arsenic content of the finished product, it can be asserted, without any fear whatever of successful contradiction, that the arsenic finds its way into the goods by way of the sulphuric acid which is used in the manufacture of all the permitted colors, as well as in the manufacture of Paranitranilin and Betanaphthol. If arsenic from this source can be kept out of these substances it can also be excluded from the permitted colors.

Turning now to those ingredients below the parallel lines, the only means of introducing arsenic would be through the hydrochloric acid used, which in turn derives its arsenic from the sulphuric acid used in its manufacture, and since arsenic free-sulphuric acid can be used in the first stages of producing the seven permitted colors and the paranitranilin and betanaphthol, it can also be employed in the manufacture of the hydrochloric acid used in the subsequent stages of manufacture of the seven permitted colors. Therefore, it seems unreasonable to permit a higher arsenic content in food colors than in paranitranilin and betanaphthol, which are sold in open competition, and which are used for the production of the very cheapest colored cloths, inks, and paints.

With reference to the content of iron, and other heavy metals, which satisfied the pharmacopæial tests in the case of paranitranilin and betanaphthol, these materials probably enter the product from the vessels in which the manufacturing operations are performed. The same kind of vessel used in the manufacture of paranitranilin

and betanaphthol is used in making the seven permitted colors, and since the heavy metals can be and are kept out of these two substances to the extent required by the Pharmacopæia, there seems to be no good reason why they should not be kept out of the seven permitted colors when made for use in food products.

Since all these defects in the seven permitted colors can be obviated in their first stage of manufacture (as is shown in the case of paranitranilin and betanaphthol, and in some actual commercial samples of 1907, see sections XIV and XV), they are commercially avoidable at subsequent stages of manufacture, and there is no good reason why they should be then introduced.

# XVII. METHODS OF ANALYSIS USED IN TESTING COLORS FOR CERTIFICATION.

# INTRODUCTION.

The exact analytical methods developed and tested in the New York Food and Drug Inspection Laboratory and used in obtaining the analytical data contained in the preceding chapter are described in the following pages. These descriptions were written by A. F. Seeker, of that laboratory, under whose immediate supervision all the laboratory work was done, and they represent a great deal of work, extending over more than three years. The methods are submitted in the hope and expectation that in their wider application by a larger number of chemists any defects in the methods or conclusions drawn from the results will be detected and rectified. Experience in the New York laboratory has shown that even different chemists of varying degrees of experience in this particular line of work obtain concordant duplicates after a comparatively short laboratory acquaintance with these methods.

The identity of these colors and their freedom from foreign dyes is shown by the close agreement of their elements, as determined by analysis, with their theoretical composition; their behavoir toward reagents as given by standard works on dyes; their distribution between the layers when neutral, alkaline, and acid aqueous solutions are shaken with various immiscible solvents; uniformity of shade in the spots produced by particles of the dry color blown over the surface of wet filter paper, or water, and over concentrated sulphuric acid; uniformity of shade produced by a 0.5 per cent dyeing on wool under standard conditions, with similar dyeings from fractions produced by partial precipitation, by partial salting out, by fractional crystallization, and by extraction with alcohol or some liquid in which the pure color is not very soluble; and the behavior of the dye in acid and alkaline solution toward cotton. Which of these tests are needed to prove conclusively the identity of certain dyes or to establish their

absence, is a matter that varies so much from case to case that it must be left largely to the individual judgment to decide on the best combination of tests, and for that reason such combinations are not here offered. The general methods and procedures just outlined have, however, when properly combined, led to satisfactory results.

In order to compare the results of the color analyses on the same basis, the actual figures obtained in the various determinations besides being reported as found, are also recalculated on a basis of 100 parts of actual color, i. e., the sum of the percentages of material which is not coloring matter, such as moisture, total insoluble matter, sodium chlorid, sodium sulphate, and ether extractives is deducted from 100 and the percentages of the various constituents found divided by the difference, the quotient being then multiplied by 100. The percentage of sodium in actual dye is calculated from the sulphated ash. The methods of analysis of the seven permitted colors of Food Inspection Decision No. 76 are given in the following order:

- 1. Naphthol Yellow S.
- 2. Ponceau 3R.
- 3. Orange I.
- 4. Amaranth.

- 5. Light Green S F Yellowish.
- 6. Erythrosin.
- 7. Indigo Disulphoacid.

#### NAPHTHOL YELLOW S.

#### MOISTURE.

Dry from 1 to 2 grams of the finely powdered dye in an air oven at 120° to 125° C. to constant weight.

#### TOTAL INSOLUBLE MATTER.

Dissolve 5 grams of color in 200 cc of hot distilled water, filter through a tared gooch, wash till the washings run through colorless, dry the insoluble residue at 105° C., and weigh.

#### NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Ignite the gooch containing the insoluble matter of the last determination at a low red heat, cool, and weigh.

# SODIUM CHLORID.

Dissolve 3 to 5 grams of dye in 200 cc of water, acidify the solution with nitric acid, and precipitate the chlorin as silver chlorid. The latter is separated, washed, ignited, and weighed in a tared gooch crucible in the usual way.

#### SODIUM SULPHATE.

Dissolve 1 gram of dye in about 100 cc of water in a 200-cc graduated flask, add 40 cc of a 20 per cent solution of potassium chlorid, shake the mixture well, make up to mark with water, shake again,

and then filter through dry paper. Treat 100 cc of the filtrate (representing 0.5 gram of dye) with 5 cc of 10 per cent barium chlorid solution without acidifying, and allow to stand overnight. If a precipitate forms, it is filtered, washed, ignited, and weighed in the usual way.

## HEAVY METALS.

Treat the sulphated ash from 1 gram of the sample with 20 cc of water, digest with 10 cc of 10 per cent hydrochloric acid till solution is complete, place 3 cc of the mixture in a test tube, add 10 cc of freshly prepared hydrogen sulphid test solution (U. S. P.), shake the mixture, warm to 50° C., stopper, and allow to stand in a warm place (at about 35° C.) for half an hour. Run a blank test at the same time, with the same amount of hydrogen sulphid solution, using water instead of the solution containing the color ash. No turbidity other than that sometimes produced by slight separation of sulphur should appear in this test. Both tubes are then made slightly alkaline with ammonium hydroxid, and no precipitate should be produced, although a slight coloration, due to the presence of a small amount of iron, sometimes occurs. If this coloration is very marked the amount of iron should be determined. This is done by digesting the sulphated ash from a weighed amount of the sample with hydrochloric acid until all of the iron has gone into solution. The solution is filtered, and the filtrate poured into an excess of hot, pure, freshly prepared sodium hydroxid (by sodium) solution in a platinum dish. The precipitate is washed, dissolved in dilute hydrochloric acid, and again precipitated with ammonium hydroxid. The last precipitate is washed, ignited, and weighed in the usual manner.

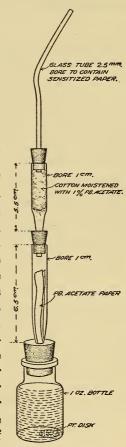
# ARSENIC (SEEKER AND SMITH'S METHOD).

Dissolve 10 grams of the dye in 200 cc of water, heating to insure complete solution of the color, add about 10 cc of strong bromin water to convert any arsenite to arsenate. Make the mixture alkaline with a few cubic centimeters of strong ammonium hydroxid. Twenty cubic centimeters of a sodium phosphate solution containing 100 grams of crystallized sodium phosphate per liter are next added from a pipette, after which magnesia mixture (containing 55 grams of hydrated magnesium chlorid, 55 grams of ammonium chlorid, and 88 cc of ammonium hydroxid, specific gravity 0.9 per liter) is added from a burette, stirring vigorously. The amount of magnesia added should be in slight excess of that necessary completely to precipitate the phosphate and should previously be ascertained by blank experiment. Then add 10 cc of ammonium hydroxid (specific gravity 0.90), and allow the whole to stand for at least three hours; separate the precipitate by filtration and wash it free, or nearly so, of

dye with ammonium hydroxid containing one-tenth its volume of ammonia (specific gravity 0.90). Dissolve the precipitate from the paper with 1:1 nitric acid, the washings being collected in a large porcelain crucible; add 5 cc of sulphuric acid to the contents of the crucible, and evaporate the whole almost to dryness. It is not necessary that the solution should be colorless at this point, a brown colored solution giving equally accurate results. Add 20 cc of water

to the residue in the crucible and then 10 cc of a saturated solution of sulphur dioxid; evaporate the solution to a sirupy consistency to remove the sulphur dioxid, and then take up in 20 cc of water and place in a 30-cc evolution bottle, add 5 cc of concentrated sulphuric acid, and determine the arsenic in the form of apparatus (fig. 1) used by Bishop in his modification of the Gutzeit test, the stains obtained being compared with those given by known amounts of arsenic.

The apparatus used by Bishop consists of the following parts: A 30-cc salt mouth evolution bottle into which is fitted a one-hole rubber stopper carrying a glass tube 6.5 cm long with an internal diameter of 1 cm, this tube in turn being provided with a one-hole rubber stopper fitted with another tube of the same diameter and 5.5 cm long, the diameters of both tubes being constricted at the points at which they are inserted in the rubber stoppers. A third glass tube 15 cm long, having an internal diameter of 2.5 mm, is fitted into the second at its upper end by means of a rubber stopper. The first tube contains a strip of filter paper which has been saturated with a 5 per cent solution of lead acetate and dried. The second tube contains a loosely packed plug of cotton-wool freshly moistened with a 1 per cent Fig. 1.-Apparatus for the solution of lead acetate. Into the topmost tube



determination of arsenic.

is inserted the strip of sensitized paper to receive the arsenic stain. The arsin is generated by introducing into the evolution bottle six pieces of Kahlbaum's stick zinc (arsenic-free for forensic purposes) weighing in all about 8 grams, and to assist in an active and constant evolution of gas a disk of platinum is also placed in each bottle to form an electrolytic couple. The evolution of gas is allowed to proceed for one hour. The stains are produced on strips of hard pressed white paper (2 mm wide and 120 mm long) that has been sensitized by being

dipped in a 5 per cent alcoholic solution of mercuric chlorid and then dried. (Note.—It has recently been found that mercuric bromid yields stains that are more evenly distributed and also produces standards that are incomparably more permanent.) For purposes of comparison it is better not to develop the strips stained by the arsin, though some prefer to dip the stains in ammonium hydroxid, which causes them to become black. A blank is run with each set of determinations.

#### ETHER EXTRACTIVES.

Dissolve 10 grams of color in 150 cc of water and extract in a separatory funnel with ether that has been washed with water (using three 150 cc portions of water for each liter of ether). Extract the color solution with two 100 cc portions of this ether, shaking thoroughly for one minute, and wash the combined ether extract successively with 35, 20, and 10 cc of water made alkaline or acid, as the case requires, with 1 cc of tenth normal alkali or acid per 100 cc of water. Decant the ether from the mouth of the separatory and rinse the funnel once with 5 cc ether. The color solution is first extracted neutral, the extracted solution being then rendered alkaline with 2 cc of a 10 per cent solution of caustic soda and again extracted with two 100-cc portions of ether. In acidifying for the third extraction, add twice the amount of hydrochloric acid (1 to 3) necessary to neutralize the alkali, and repeat the extraction with two 100-cc portions of ether. Place the neutral, alkaline, and acid extracts in a dust-free atmosphere, and allow the ether to evaporate spontaneously, after which dry the residues to constant weigh over sulphuric acid, using flat-bottomed dishes 23 inches in diameter, 11 inches in height, and of about 100 cc capacity. The dishes should be thoroughly cleaned, wiped dry, and allowed to stand in a sulphuric acid desiccator at least two hours before weighing. In order to avoid the generation of static charges of electricity, they should not be wiped immediately before weighing. Run two blank determinations with each series of ether extracts, and deduct the average gain in weight of these two blanks from the weights obtained in the other determinations.

#### SULPHATED ASH.

Weigh accurately 0.5 to 1 gram of the color into a wide platinum dish, moisten with concentrated sulphuric acid, and ignite cautiously, avoiding spattering; moisten the residue repeatedly with sulphuric acid and ignite until all the carbon is removed and a white or reddish ash is obtained. This is finally ignited at a fairly bright red heat, cooled, and weighed. The aqueous solution of this ash should be neutral to litmus, and may be used in a quantitative test for potassium.

#### CALCIUM.

Digest the residue from the sulphated ash determination with hydrochloric acid, render the solution alkaline with ammonium hydroxid, filter, and precipitate the calcium in the filtrate with ammonium oxalate. The precipitate of calcium oxalate is filtered on a tared gooch, washed, dried at 100° C. and weighed as calcium oxalate, the calcium being calculated from the formula CaC<sub>2</sub>O<sub>4</sub>.H<sub>2</sub>O. This method is applicable only in cases where the amount of calcium is very small. When the amount exceeds 0.5 per cent it should be determined by digesting the sulphated ash with ammonium sulphate solution made acid with hydrochloric acid, and precipitating the iron by an excess of ammonium hydroxid. The precipitate is washed, dissolved in hydrochloric acid, and reprecipitated, the filtrate and washings being added to those obtained from the first precipitate. The calcium is precipitated in the combined filtrates with ammonium oxalate, the oxalate being filtered, washed, ignited, and weighed in the usual way; the residue is weighed as oxid after ignition to bright redness.

#### SULPHUR.

Determine upon 0.2 to 0.3 gram portions by the Carius method (Gattermann, Practical Methods of Organic Chemistry, 1901, p. 81), using 3 cc of fuming nitric acid (sp. gr. 1.5), and heating the sealed tubes to 300° C. for at least eight hours.

#### NITROGEN.

Use the method of Dumas (Gattermann, Practical Methods of Organic Chemistry, 1901, p. 85).

#### PONCEAU 3R.

#### MOISTURE.

Dry 1 to 2 grams of the finely ground dye at 109° to 110° C. in a current of dry hydrogen to constant weight.

# TOTAL INSOLUBLE MATTER.

Determine as under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Determine as under Naphthol Yellow S, page 211.

SODIUM CHLORID (SEEKER AND MATHEWSON'S METHOD).

Mix 2 grams of dye thoroughly with from 2 to 3 grams of sodium carbonate, moisten with water to form a paste, again mix, dry, and ignite at a low red heat. By moistening, drying, and reigniting a

more complete destruction of organic matter is obtained. Break up the charred mass, introduce into a 200-cc graduated flask with about 100 to 150 cc of hot water, and add an excess of potassium permanganate to oxidize sulphids. Destroy the excess of permanganate by adding sulphur dioxid solution until the red color changes to brown, then cool the mixture and make up to the mark with water. Filter through a dry paper, acidify 100 cc of the filtrate with nitric acid, and precipitate the chlorin as silver chlorid by the addition of silver nitrate. If the solution should be brownish from a trace of organic matter the silver chlorid does not readily coagulate and tends to pass through the filter. In this case a few drops of potassium permanganate solution are added to this acid mixture, the organic matter being almost instantly oxidized. The mixture is then decolorized with a few drops of sulphur dioxid solution, the silver chlorid is separated on a tared gooch, washed, ignited, and weighed in the usual manner.

The following method may also be used for Ponceau 3R:

Dissolve 5 grams of the dye in 15°C cc hot water, wash into a 25°C-cc graduated flask, and add 25°C-cc of a 10°P-cent solution of barium nitrate. Cool the mixture, make up to the mark, and filter through a dry paper; acidify 10°C-cc of the filtrate, representing 2 grams of color, with nitric acid, and treat with silver nitrate solution, the precipitated silver chlorid being separated, washed, ignited, and weighed in a tared gooch crucible in the usual way.

# SODIUM SULPHATE.

Dissolve 2 grams of dye in 100 cc of hot water, wash into a 200-cc graduated flask, add 50 grams of pure sodium chlorid, cool, and make up to the mark with water. Filter through a dry filter, dilute 100 cc of the filtrate to 300 cc, acidify with hydrochloric acid and precipitate the sulphates with barium chlorid. Filter, wash, ignite, and weigh in a tared platinum gooch in the usual way.

# HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

#### ARSENIC.

Determine as given under Naphthol Yellow S, page 212.

#### ETHER EXTRACTIVES.

Determine as given under Naphthol Yellow S, page 214.

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

#### CALCIUM.

Determine as given under Naphthol Yellow S, page 215.

#### SULPHUR.

Determine as given under Naphthol Yellow S, page 215.

NITROGEN (SEEKER AND MATHEWSON'S METHOD).

Treat 2 grams of the color with 25 cc of a saturated solution of sulphur dioxid and 1 gram of zinc dust and warm the mixture gently until it becomes colorless. This should take place in from two to three minutes, but if it does not add more sulphur dioxid solution in small portions at a time until the color is destroyed. Then add 30 cc of concentrated sulphuric acid and 0.7 gram of mercuric oxid or its equivalent of metallic mercury and digest the mixture, make alkaline, and distil as directed on page 6, Bulletin 107, Revised, Bureau of Chemistry, United States Department of Agriculture, under the Kjeldahl process.

## CRUDE CUMIDIN.

Dissolve 20 grams of dye in 400 cc of hot water and pour the solution, a little at a time, into a reducing solution composed of 75 grams of stannous chlorid dissolved in 180 cc of concentrated hydrochloric acid. Heat the mixture on a steam bath until it is straw colored, cool, add an excess of sodium hydroxid, and extract in a separatory funnel with ether. Separate the ether layer and distil off the solvent until the residue measures about 50 cc. Then cautiously heat over the steam with constant agitation until the odor of ether disappears, after which the last of the moisture is removed by introducing a few pieces of solid caustic soda and allowing to stand. The residue consists of crude cumidin and should boil above 220° C. Cumidin nitrate is sparingly soluble in water.

#### ORANGE I.

# MOISTURE.

Determine as given under Ponceau 3R, page 215.

# TOTAL INSOLUBLE MATTER

Determine as given under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

#### SODIUM CHLORID.

Determine as given under Ponceau 3R, page 215.

#### SODIUM SULPHATE.

Dissolve 1 gram of dye in 100 cc water contained in a 200-cc graduated flask and treat the solution with 60 cc of a 20 per cent solution of potassium chlorid. Make the mixture up to the mark with water, shake, filter through a dry paper, dilute an aliquot of 100 cc of the filtrate to 200 cc, acidify with 1 cc of 10 per cent hydrochloric acid, treat with 5 cc of 10 per cent barium chlorid solution, and allow to stand over night. If a precipitate has been formed this is separated, ignited, and weighed in the usual way.

# HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

ARSENIC (SEEKER AND SMITH'S METHOD).

Dissolve 2 grams of dye in a mixture of 130 cc water and 70 cc of 95 per cent alcohol. Add about 10 cc of strong bromin water to convert any arsenite to arsenate. Make the mixture alkaline with a few cubic centimeters of strong ammonium hydroxid and add, from a pipette, 20 cc of a sodium phosphate solution containing 100 grams of crystallized sodium phosphate per liter, after which magnesia mixture (containing 55 grams of hydrated magnesium chlorid. 55 grams of ammonium chlorid, and 88 cc of ammonium hydroxid, sp. gr. 0.9, per liter) is added from a burette, stirring vigorously. The amount of magnesia mixture to be added should be in slight excess of that necessary to precipitate the phosphate completely, and should be previously ascertained by a blank experiment. Finally add 10 cc of ammonium hydroxid (sp. gr. 0.96) and allow the whole to stand for at least eight hours. Separate the precipitate by filtration and wash it free, or nearly so, of dye with a mixture of one-third alcohol and two-thirds water containing one-tenth its volume of ammonium hydroxid (sp. gr. 0.90). Dissolve the precipitate from the paper with 20 per cent sulphuric acid, the washings being collected in a large porcelain crucible. Add 5 cc of concentrated nitric acid to the contents of the crucible and evaporate the whole almost to dryness. The mixture need not be colorless at this point, a brown colored solution giving equally accurate results. Add 20 cc of water to the residue in the crucible and then 10 cc of a saturated solution of sulphur dioxid. Evaporate the solution to a syrupy consistency to remove sulphur dioxid, and then take up in 20 cc of water, place in a 30-cc evolution bottle, add 5 cc of concentrated sulphuric acid, and determine the arsenic by the modified Gutzeit method as given under Naphthol Yellow S, page 213.

# ETHER EXTRACTIVES.

Determine as given under Naphthol Yellow S, page 214

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

CALCIUM.

Determine as given under Naphthol Yellow S, page 215.

SULPHUR.

Determine as given under Naphthol Yellow S, page 215.

NITROGEN.

Determine as given under Ponceau 3R, page 217.

TEST FOR ORANGE II (SMITH AND MATHEWSON'S METHOD).

The following solutions are required:

- (1) Fifteen per cent titanium trichlorid.
- (2) Freshly prepared diazotized sulphanilic acid. Heat a mixture composed of 1 gram of sulphanilic acid, 10 cc of water, and 20 cc of concentrated hydrochloric acid on a steam bath for five minutes with occasional shaking. Cool to about 10° C. and add slowly 10 cc of a 1 per cent solution of sodium nitrite. Allow the mixture to stand at about 10° for 10 minutes, dilute to 1 liter, and shake until all the solid particles have dissolved.
- (3) Stannous chlorid, prepared by dissolving 40 grams of stannous chlorid in 100 cc of concentrated hydrochloric acid. Dilute this ten times with water, immediately before using.

Place 2 cc of a 0.1 per cent solution of the dye in a colorimeter tube having a capacity of about 100 cc, add a small drop of titanium trichlorid solution and shake until the mixture is decolorized. Standards containing the same quantity of color composed of a mixture of pure Orange I and known amounts of Orange II are treated in the same way, the volume of the solution at this point to measureless than 5 cc. Dilute the decolorized solutions to 50 cc with 95 per cent alcohol and equalize the temperature by immersing the tubes in water at room temperature. (Note.—A slight coloration that may develop at this point may be disregarded.) Allow the tubes to stand in the water for about five minutes, add 2 cc of the diazotized sulphanilic acid, and mix thoroughly. If the titanium trichlorid has not been used in too great excess, the first few drops of the diazotized sulphanilic acid will cause a formation of color. Allow the coupling to proceed for three minutes and then add 5 cc of the diluted stannous chlorid with vigorous shaking. In two minutes the blue color due to Orange I will disappear, leaving only the pink caused by the coupling product of Orange II. The depth of color in the tube containing the dve under examination may then be compared to the standards.

#### AMARANTH.

#### MOISTURE.

Determine as given under Ponceau 3R, page 215.

#### TOTAL INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

#### SODIUM CHLORID.

Determine as given under Ponceau 3R, page 215.

#### SODIUM SULPHATE.

Dissolve 2 grams of dye in 100 cc of warm water in a 200-cc graduated flask, and add 36 grams of pure sodium chlorid. Allow the mixture to stand with frequent shaking for one hour, and after cooling make up to the mark with a saturated salt solution. Shake the mixture and filter through a dry paper; dilute 100 cc of the filtrate (representing 1 gram) with water, acidify with hydrochloric acid, and precipitate the sulphates with barium chlorid. The precipitate is separated, washed, and ignited upon a tared platinum gooch crucible.

# HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

#### ARSENIC.

Determine as given under Naphthol Yellow S, page 212.

# ETHER EXTRACTIVES.

Determine as given under Naphthol Yellow S, page 214.

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

#### CALCIUM.

Determine as given under Naphthol Yellow S, page 215.

#### SULPHUR.

Determine as given under Naphthol Yellow S, page 215.

#### NITROGEN.

Determine as given under Ponceau 3R, page 217.

## LIGHT GREEN S F YELLOWISH.

#### MOISTURE.

Determine as given under Ponceau 3R, page 215.

TOTAL INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

#### SODIUM CHLORID.

Determine as given under Ponceau 3R, page 215.

#### SODIUM SULPHATE.

Dissolve 5 grams of the color in 100 cc of water, warming by means of a gentle heat. Dissolve 11 grams of safranin in a separate portion of 400 cc of water, also by warming, taking care in both instances to prevent loss by evaporation. Mix the two solutions, shake thoroughly, and filter through a dry filter. Render an aliquot portion of the filtrate alkaline with sodium hydroxid and remove the excess of safranin by shaking with two successive portions of amyl alcohol. Wash the combined amyl alcohol layers with two portions of water and add the washings to the main aqueous solution, which is then acidified with hydrochloric acid and sulphates determined in the usual manner by precipitation with barium chlorid as barium sulphate.

#### HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

#### ARSENIC.

Determine as given under Naphthol Yellow S, page 212.

#### ETHER EXTRACTIVES.

Determine as given under Naphthol Yellow S, page 214

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

# CALCIUM.

Determine as given under Naphthol Yellow S, page 215

#### SULPHUR.

Determine as given under Naphthol Yellow S, page 215.

#### NITROGEN.

Determine on 2-gram portions by Gunning's modification of the Kjeldahl process, using a little copper sulphate to assist the oxidation (see page 7, Bulletin 107, Revised, Bureau of Chemistry, United States Department of Agriculture).

# ERYTHROSIN.

#### MOISTURE.

Determine as given under Ponceau 3R, page 215.

#### TOTAL INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER

Determine as given under Naphthol Yellow S, page 211.

#### SODIUM CHLORID.

Dissolve 5 grams of the dye in 400 cc water and acidify with dilute nitric acid. Make the mixture up to 500 cc with water, and then filter through a dry filter. Determine chlorids in an aliquot of 200 cc of the filtrate by precipitation with silver nitrate, washing, igniting, and weighing the silver chlorid in a tared gooch crucible in the usual manner.

#### SODIUM SULPHATE.

Employ another aliquot of the filtrate obtained after precipitating the color acid as above in the determination of sulphates, precipitating the latter as barium sulphate in the usual manner.

#### HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

ARSENIC (SEEKER AND SMITH'S METHOD).

Dissolve 16 grams of dye in 370 cc of water, add 5 cc of strong bromin water, and finally 25 cc of dilute sulphuric acid (1 to 4). Shake thoroughly and filter through a dry filter. Place an aliquot of 250 cc, representing 10 grams of color from the filtrate, in a porcelain casserole, add 5 cc concentrated nitric acid (very important to prevent loss of arsenic), and evaporate till fuming has ceased. Reduce the residue with sulphur dioxid solution, evaporate to small bulk, and determine the arsenic in the form of apparatus used by Bishop in

his modification of the Gutzeit test. See under Naphthol Yellow S, pages 212 and 213. (Note.—It is somewhat difficult at times to recover 250 cc of filtrate, but less may be used and a correction made, if necessary.)

#### ETHER EXTRACTIVES.

Determine as given under Naphthol Yellow S, page 214, omitting the acid extraction.

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

IODIN, BROMIN, AND CHLORIN (CORNELISON'S METHOD).

Mix 0.2 to 0.3 gram of the sample with 2 grams of pure potassium bichromate and 15 cc of strong sulphuric acid in the evolution flask of an apparatus made entirely of glass, with ground-glass joints. Thoroughly mix the contents of the evolution flask, so that all lumps are disintegrated, and then heat at 100° C. for 15 minutes, after which raise the temperature to 150° C. for thirty minutes, a current of air dried over calcium chlorid and potassium hydroxid being drawn through the apparatus during this time. Iodin remains in the evolution flask as iodic acid; bromin passes off as such, and may be absorbed by allowing the air passing through the apparatus to bubble through 1 per cent sodium hydroxid. Chlorin passes out of the evolution flask as chromyl chlorid, and may also be absorbed in sodium hydroxid. Cool the mixture containing the iodic acid, and reduce the chromic acid by addition of sulphur dioxid, about 20 cc of a saturated solution being required. When enough has been added, the precipitated iodin redissolves, and the clear green color of chrome alum appears. Filter, wash the paper with distilled water, dilute the filtrate and washings to about 300 cc, and add an excess of silver nitrate. Boil till the silver iodid has flocculated, allow to stand for a few hours, and separate and weigh the silver iodid in a tared

It sometimes happens that the mixture containing the iodic acid, after the reduction with sulphur dioxid, becomes turbid, owing, apparently, to separation of a basic chromium sulphate. Very often the turbidity can not be removed by filtering, and it has been found advisable in this case to reject the determination and begin anew.

# IODIN (SEEKER AND MATHEWSON'S METHOD).1

Place from 0.3 to 0.4 gram of the erythrosin in a porcelain casserol, dissolve this in 5 cc of a 10 per cent sodium hydroxid solution, then add 35 cc of a 7 per cent solution of potassium permanganate. After

<sup>&</sup>lt;sup>1</sup> U. S. Dept. Agr., Bureau of Chemistry, Circular 65. The estimation of iodin in organic compounds and its separation from other halogens.

mixing, cover the vessel with a watch crystal, and add 10 cc of nitric acid, keeping the dish covered. Agitate the mixture, place on a steam bath, and keep covered until spattering ceases, after which remove the watch glass and allow evaporation to proceed to dryness.¹ Treat the residue with 5 cc of 7 per cent potassium permanganate and 5 cc of concentrated nitric acid and again evaporate to dryness. Then add about 50 cc of water and 5 cc of concentrated nitric acid to the residue, following this by 40 cc of a saturated solution of sulphur dioxid, and allow the whole to stand with occasional stirring (breaking up the lumps with a glass rod) until the hydrated oxid of manganese has dissolved. Filter, and wash the paper with water. Add an excess of silver nitrate to the filtrate, and boil until sulphur dioxid has been expelled and the silver iodid has flocculated. Separate, wash, and weigh the precipitate in the usual manner.

## TOTAL HALOGENS.

Mix 0.5 to 1 gram of the dye with 4 grams of potassium carbonate, moisten to a paste, again thoroughly mix, cover with a layer of dry potassium carbonate, dry, and ignite at a low red heat. Break up the char thoroughly, digest with about 200 cc water, and filter. Wash the insoluble matter until the washings no longer react with silver nitrate; then acidify the filter and washings with nitric acid and precipitate the halogens in the usual way as silver salts.

#### INDIGO DISULPHOACID.

#### MOISTURE.

Determine as given under Ponceau 3R, page 215.

# TOTAL INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

NONVOLATILE OR INORGANIC INSOLUBLE MATTER.

Determine as given under Naphthol Yellow S, page 211.

#### SODIUM CHLORID.

Determine as given under Ponceau 3R, page 215.

#### SODIUM SULPHATE.

Dissolve 2 grams of the dye in about 160 cc of water and treat the solution with 40 grams of pure sodium chlorid. After the salt has dissolved, make up the volume to exactly 200 cc. Shake the mix-

<sup>1</sup> In the operation of drying particular care should be obserred to prevent access of reducing gases to the mixture.

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ture thoroughly and filter through a dry filter. Dilute 50 cc of the filtrate with 200 cc of water, acidify with 1 cc of 10 per cent hydrochloric acid, and treat with an excess of barium chlorid solution. After standing overnight, the precipitate is separated, washed, ignited, and weighed in the usual way.

#### HEAVY METALS.

Determine as given under Naphthol Yellow S, page 212.

ARSENIC (SEEKER AND SMITH'S METHOD).

Treat 10 grams of dye in a Kjeldahl flask with 100 cc water and 10 cc concentrated nitric acid. Warm gently and finally boil until all action has ceased. Transfer to a beaker, make alkaline with ammonium hydroxid, and proceed from this point, as in the case of the solution of Naphthol Yellow S, page 212, continuing from the point at which sodium phosphate is added.

# ETHER EXTRACTIVES.

Dissolve 3 grams of dye in 200 cc of water and extract with ether, as directed under Naphthol Yellow S, page 214.

#### SULPHATED ASH.

Determine as given under Naphthol Yellow S, page 214.

CALCIUM.

Determine as given under Naphthol Yellow S, page 215.

SULPHUR.

Determine as given under Naphthol Yellow S, page 215.

NITROGEN.

Determine as given under Light Green SF Yellowish, page 222.

# XVIII. ADDENDA.

# ADDITIONAL EXAMINATION OF COAL-TAR DYES.

Since the foregoing report was written the chemical examination of the coal-tar dye specimens collected in the summer of 1907 has been completed. This investigation shows that 7 of the Green Table numbers then reported were not on the market, although those furnishing the specimens included them at the time that the samples were supplied. These numbers are as follows: not physiologically examined,

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G. T. 49, 60, 104, 518, 523; physiologically examined, with unfavorable result, G. T. 516; with favorable result, G. T. 520.

The chemical examination also shows that each of 9 additional Green Table numbers occurred once among the unreported numbers. They are as follows: not examined physiologically, G. T. 20, 328; examined physiologically: with unfavorable results, G. T. 602; with favorable results, G. T. 92, 477, 521; with contradictory results, G. T. 16, 43, 163. G. T. 20 is anilin azo (1:8) dioxy naphthalene (3:6) disulpho acid. G. T. 328 is diamino-stilbene-disulpho acid-disazophenol.

This chemical examination finally shows that beside the Green Table numbers there occurred, once each, the following: sulphonated Victoria Blue B and a sulphonated rhodulin and twice, amido-azo-

toluene-azo-alpha-naphthol.

It is clear that this revised statement of facts is without influence on the 7 permitted dyes selected; its only effect is to add G. T. 92 (yellow), 477 (blue), and 521 (red) (each occurring but once) to, and to subtract 520 (red) (said to occur twice) from, the 16 dyes (page 166) from among which the 7 permitted dyes were selected, thus giving 18 instead of 16 dyes to choose from; however, the final selection is wholly unaffected thereby. It is also clear that any and all intermediate conclusions or comparisons are only slightly and immaterially affected, if at all, by the results of this additional examination.

# SUPPLEMENTARY LIST OF TRADE NAMES OF COAL-TAR COLORS.1

Acid Yellow S (4).
Alkali Blue (477).
Azo Acid Rubin 2B (107).
Azo Yellow (92).
Crocein Scarlet O extra (164).
Dimethylanilin Orange (87).
Eosin, water soluble (512).
Fast Red A (102).
Fast Red B (65).
Fast Red E (105).
Fast Yellow Y (8).
Indian Yellow (92).

Indulin R and B (601).
Leucindophenol (572).
Methyl Orange (87).
Naphthylamin Pink (614).
Naphthylene Blue R in crystals (639).
Soluble Blue 8B and 10B (479).
Sulphur Yellow (4).
Thiochromogen (659).
Victoria Blue (488).
Wool Black (166).
Yellow T (84).

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